11th International Conference on Inelastic X-ray Scattering (IXS2019)

IXS Opportunities offered by High Repetition Rate X-ray Free Electron Lasers

The Road to a high rep. rate x-ray FEL

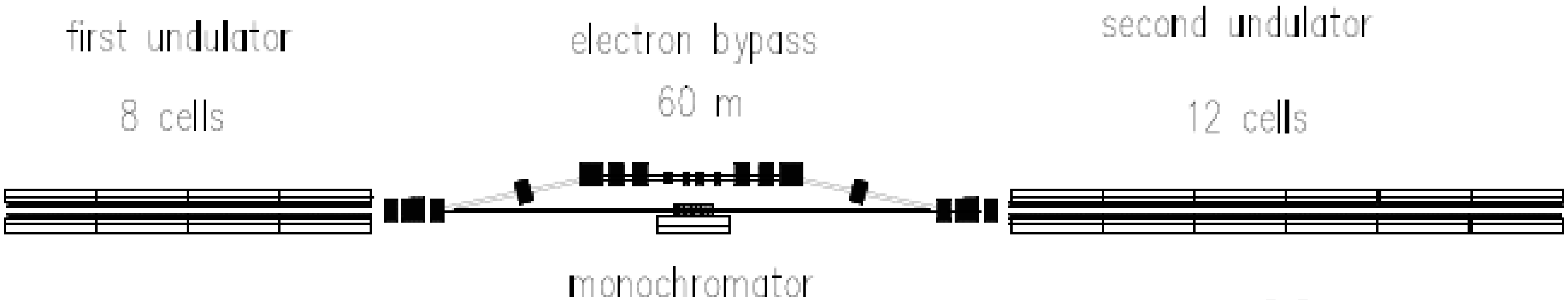
Chi-Chang Kao

June 28th, 2019

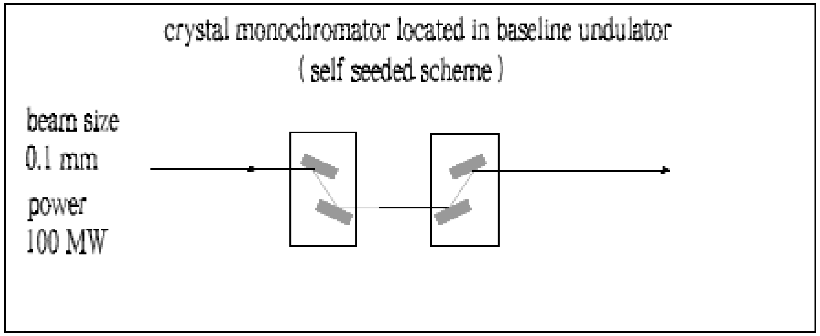
U.S. DEPARTMENT OF ENERGY
Office of Science

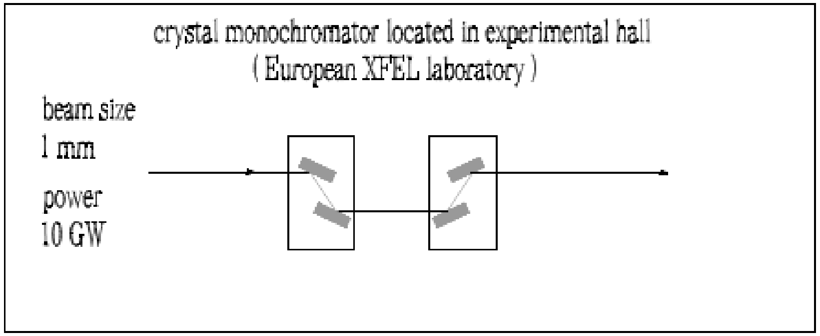
SLAC NATIONAL ACCELERATOR LABORATORY

It all started with the idea of self-seeding

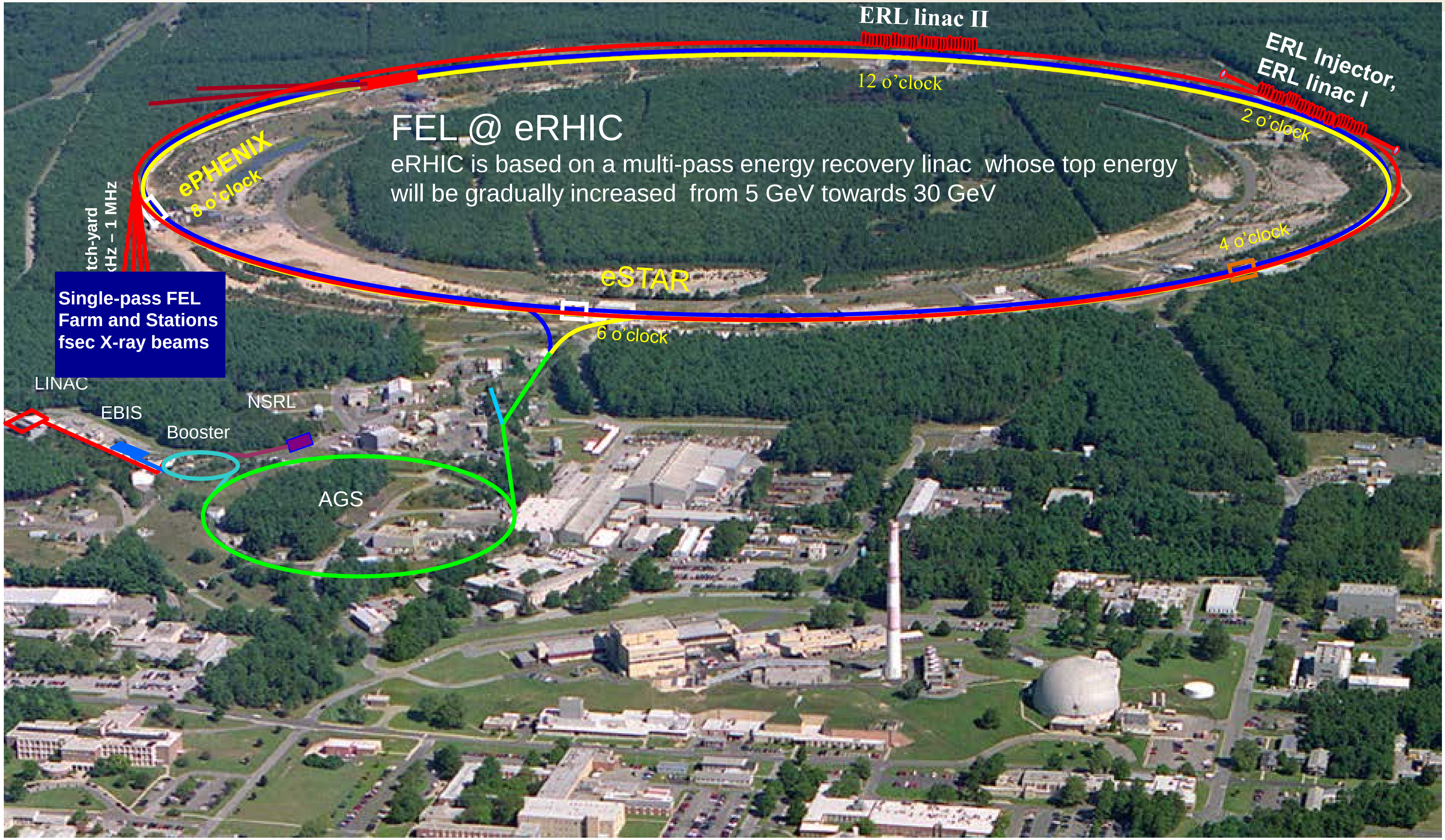


Saldin (2010)





And, here at BNL : FEL @ eRHIC (circa 2010)

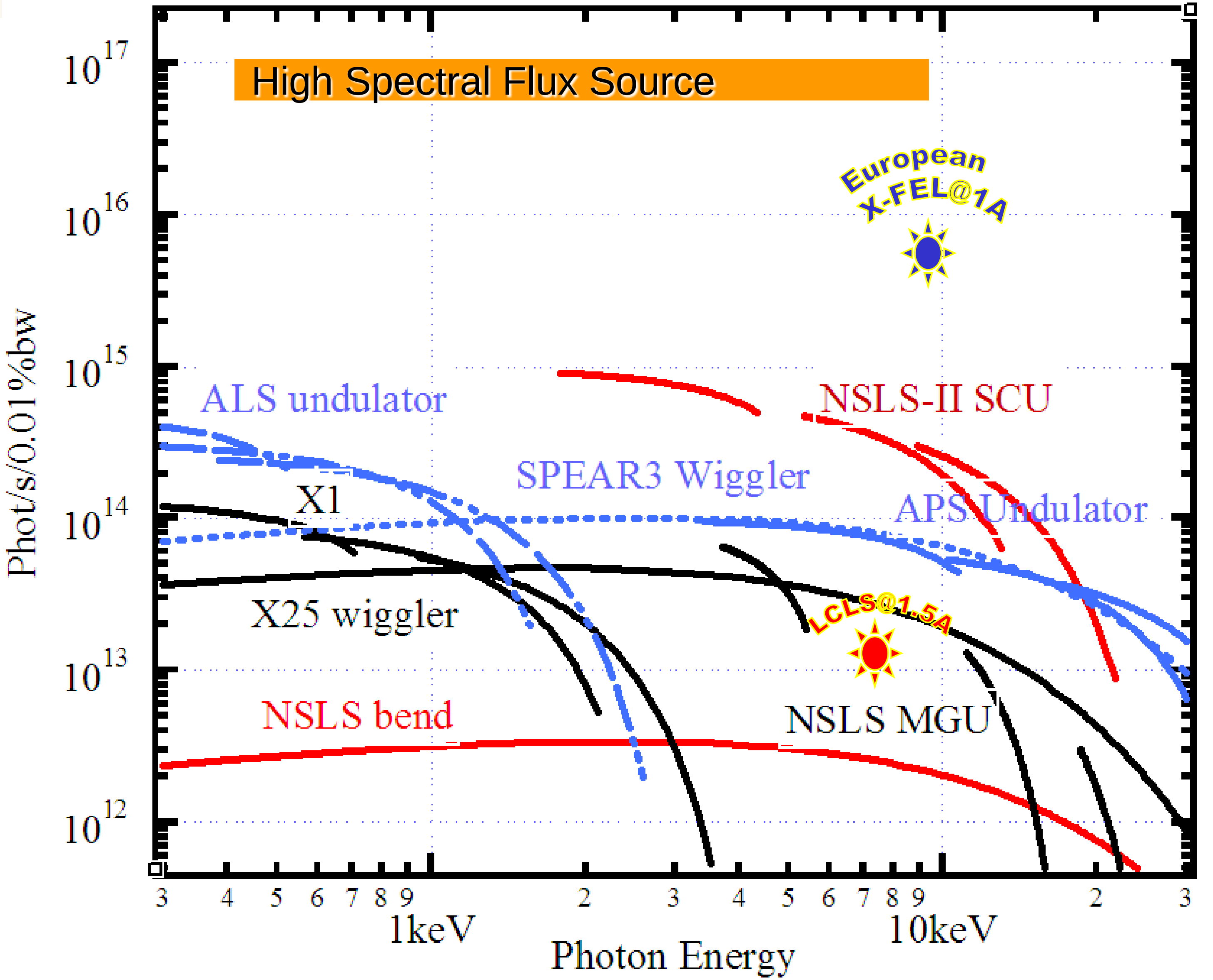


FEL @ eRHIC

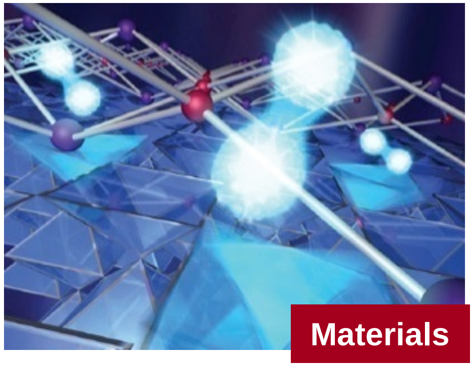
eRHIC is based on a multi-pass energy recovery linac whose top energy will be gradually increased from 5 GeV towards 30 GeV

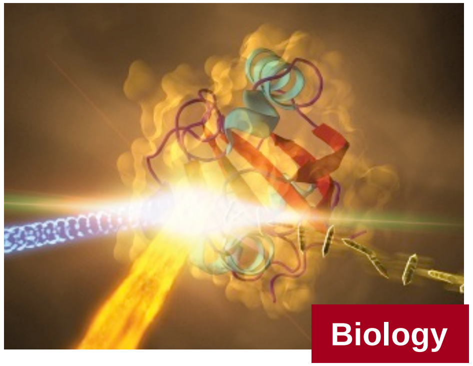
Single-pass FEL Farm and Stations fsec X-ray beams

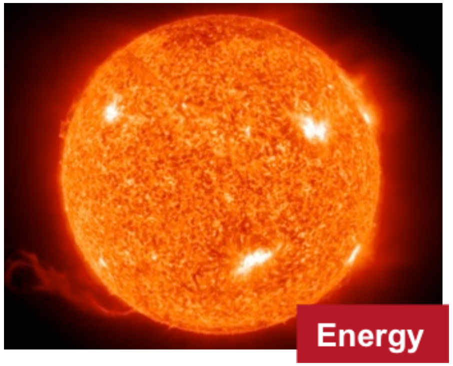
Think about future x-ray sources in terms of spectral brightness, and a factor of 1000 gain



LCLC turned on successfully in April 2009 at SLAC, with extraordinary peak power as predicted

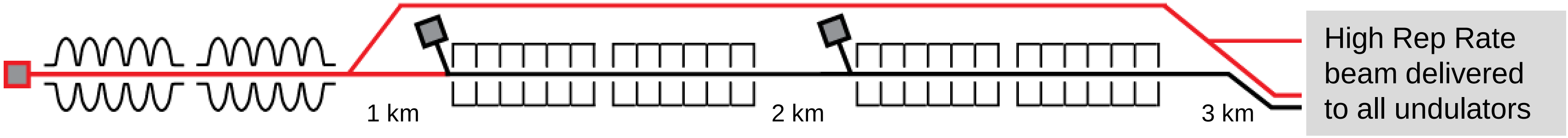




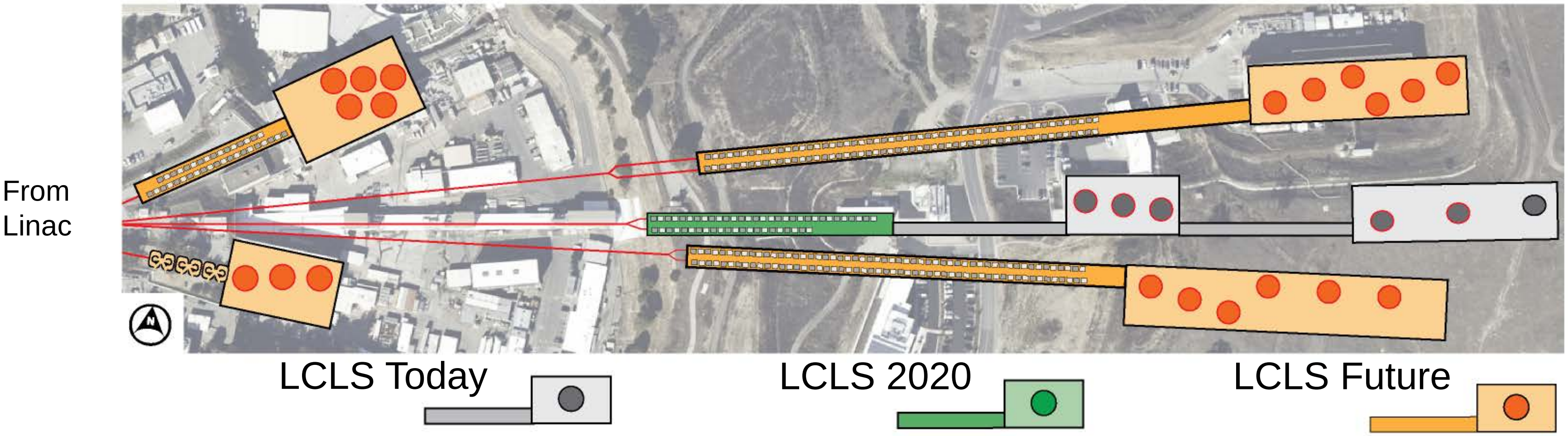


A new vision for Linac Coherent Light Source (circa 2013)

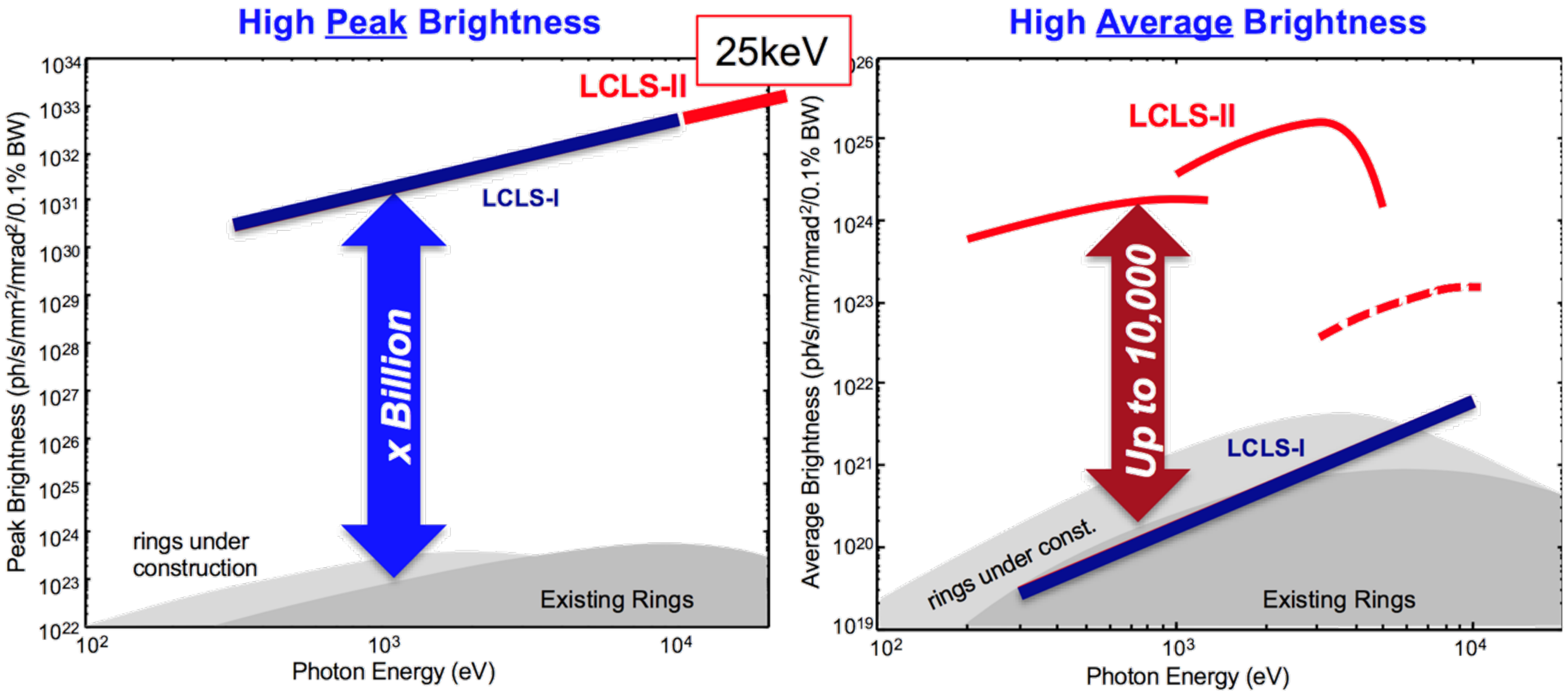
- LCLS-II project has been re-scoped to include a superconducting linac to enable MHz repetition rate
- LCLS will be *the* x-ray free electron laser center of US, which will lead to significant investment for future expansion, and great scientific opportunities for SLAC and Stanford in the coming decades



High Rep Rate beam delivered to all undulators



The first step is LCLS-II

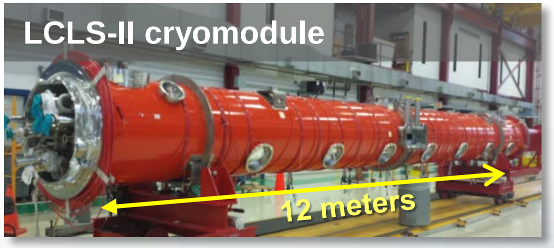


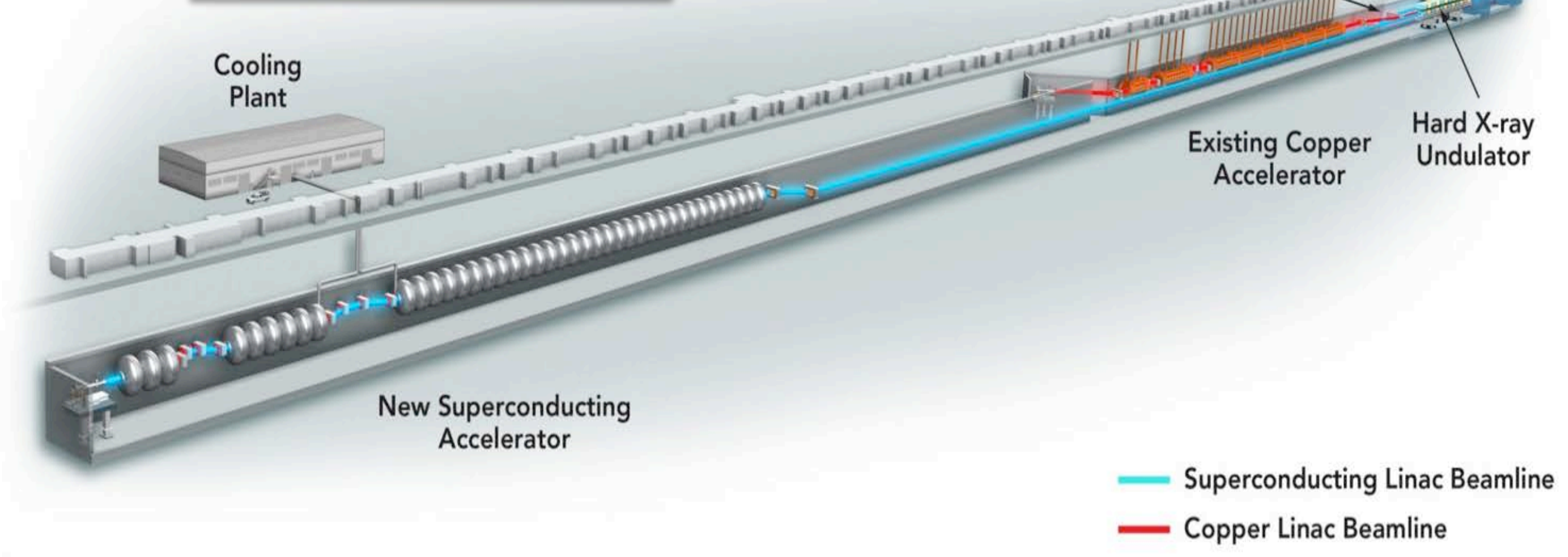
High Peak Brightness

High Average Brightness

	Now	HXU - Cu	SXU – Cu	HXU - SC	SXU - SC
Photon Energy Range (keV)	0.25 -12.8	1 - 25	0.25 - 6	1 - 5	0.25 - 1.6
Repetition Rate (Hz)	120	120	120	929,000	929,000
Per Pulse Energy (mJ)	~ 4	~ 4	~ 8	~ 0.2	~ 1
Photons/Second	~ 10^{14}	~ 10^{14}	~ 10^{14}	~ 10^{16}	~ 10^{17}

Get it done as fast as we can with a multi-lab partnership





Argonne NATIONAL LABORATORY

BERKELEY LAB Lawrence Berkeley National Laboratory

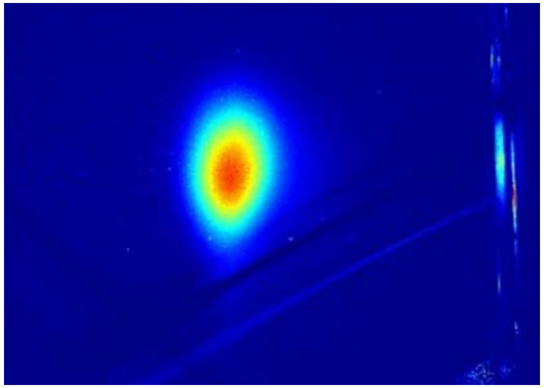
CORNELL UNIVERSITY FOUNDED A.D. 1865

Fermilab

Jefferson Lab

LCLS-II is on track: project is ~85% complete

Gun after install and vacuum connections



10 of 35 CMs installed in SLAC tunnel

Cryomodule welding at SLAC

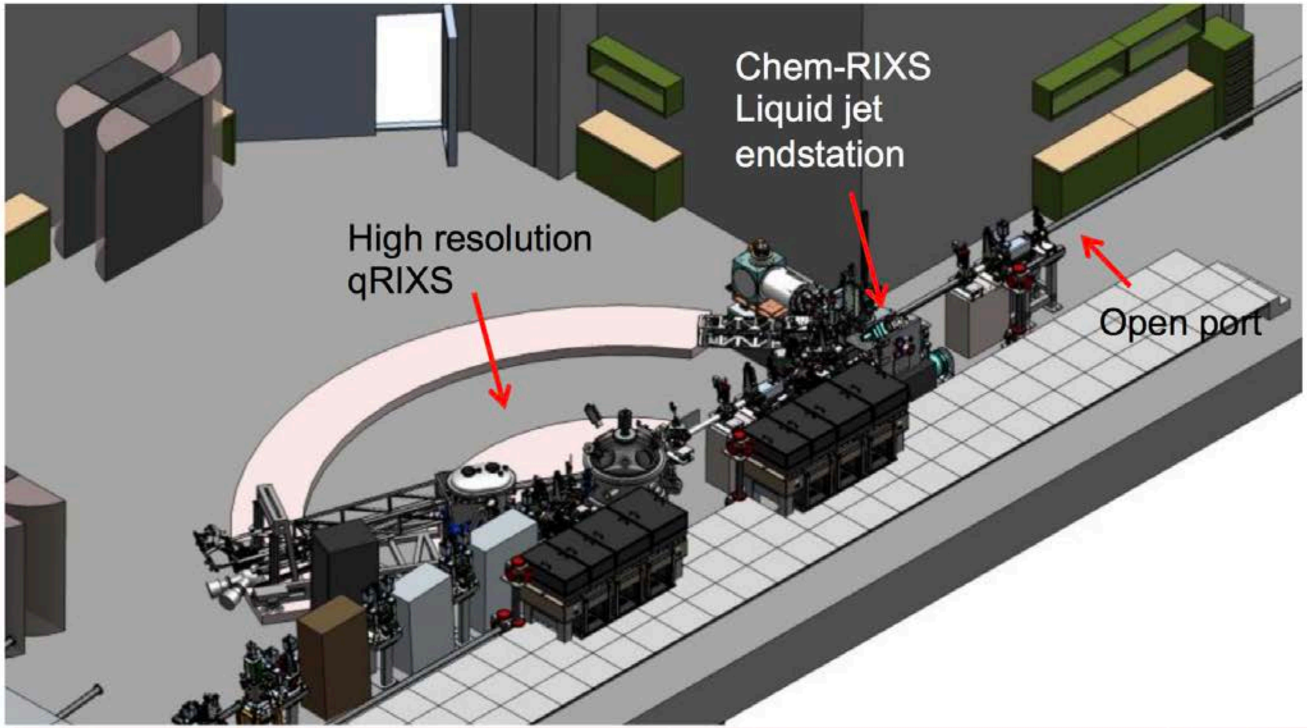
Cryoplant tank farm at SLAC

Upper cold boxes and vaporizers

2K transfer line in SLAC tunnel

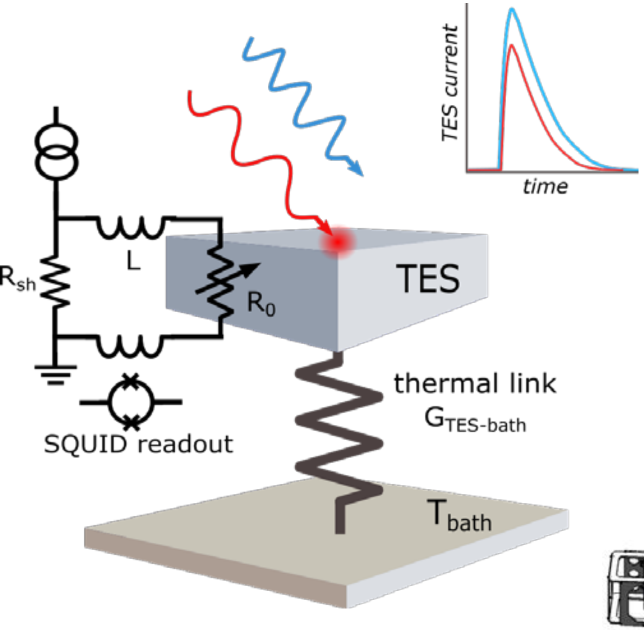
Undulator production at LBNL

RIXS is one of an the first instruments



Endstation	Science	Methods	XFEL
qRIXS	Emergent phenomena and collective modes in correlated materials	Resonant inelastic X-ray scattering, Resonant diffraction	250 – 1600 eV, $>10^{14}$ ph/s with >30,000 resolving power
ChemRIXS	Heterogeneous catalysis Interfacial chemistry Photo-catalysis	X-ray Absorption & Emission Spectroscopy	250-1600 eV, ≥ 100 kHz, 1000-5000 res. power
qRIXS	Nanoscale material dynamics	XPCS	250 – 1600 eV

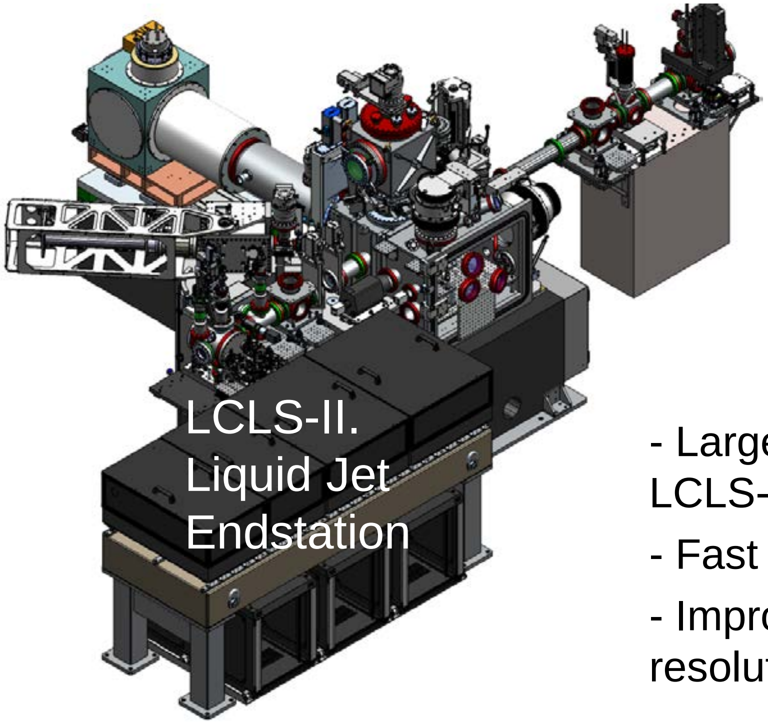
A unique opportunity: transition edge sensor (TES)



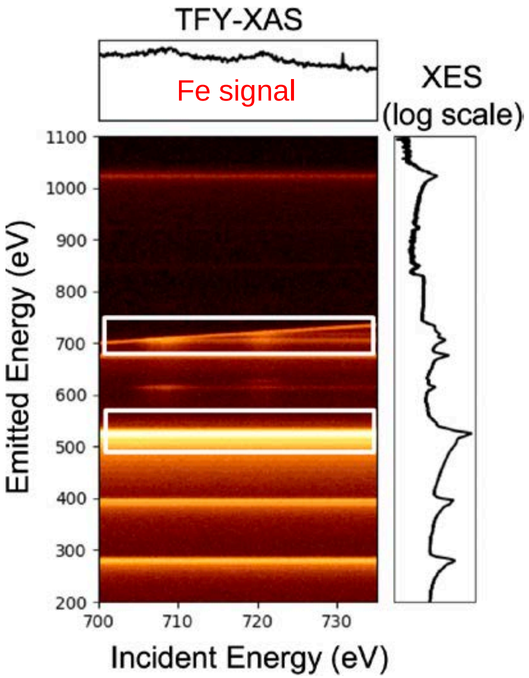
- Extremely sensitive to small energy
- Nearly 100% quantum efficiency
- Scalable to large array
- Broadband spectral coverage

Advantage:
Dilute system in Chemistry/Bio

Example: Frozen hemoglobin
(demonstrated at SSRL/SLAC)



LCLS-II.
Liquid Jet
Endstation



- Large 1000-pixel TES array for LCLS-II
- Fast readout electronics
- Improving the energy resolution (up to 0.5 eV)

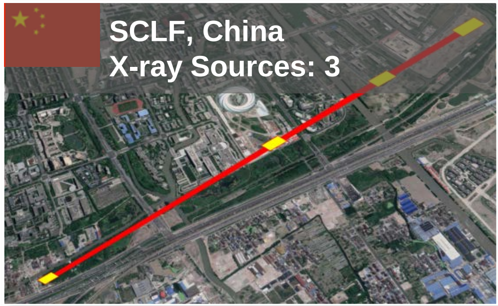
Success of LCLS has led to rapid growth in X-FELs

EuXFEL, Germany
X-ray Sources: 3 (5)

SACLA, Japan
X-ray Sources: 2 (3)

SwissFEL, Switzerland
X-ray Sources: 2

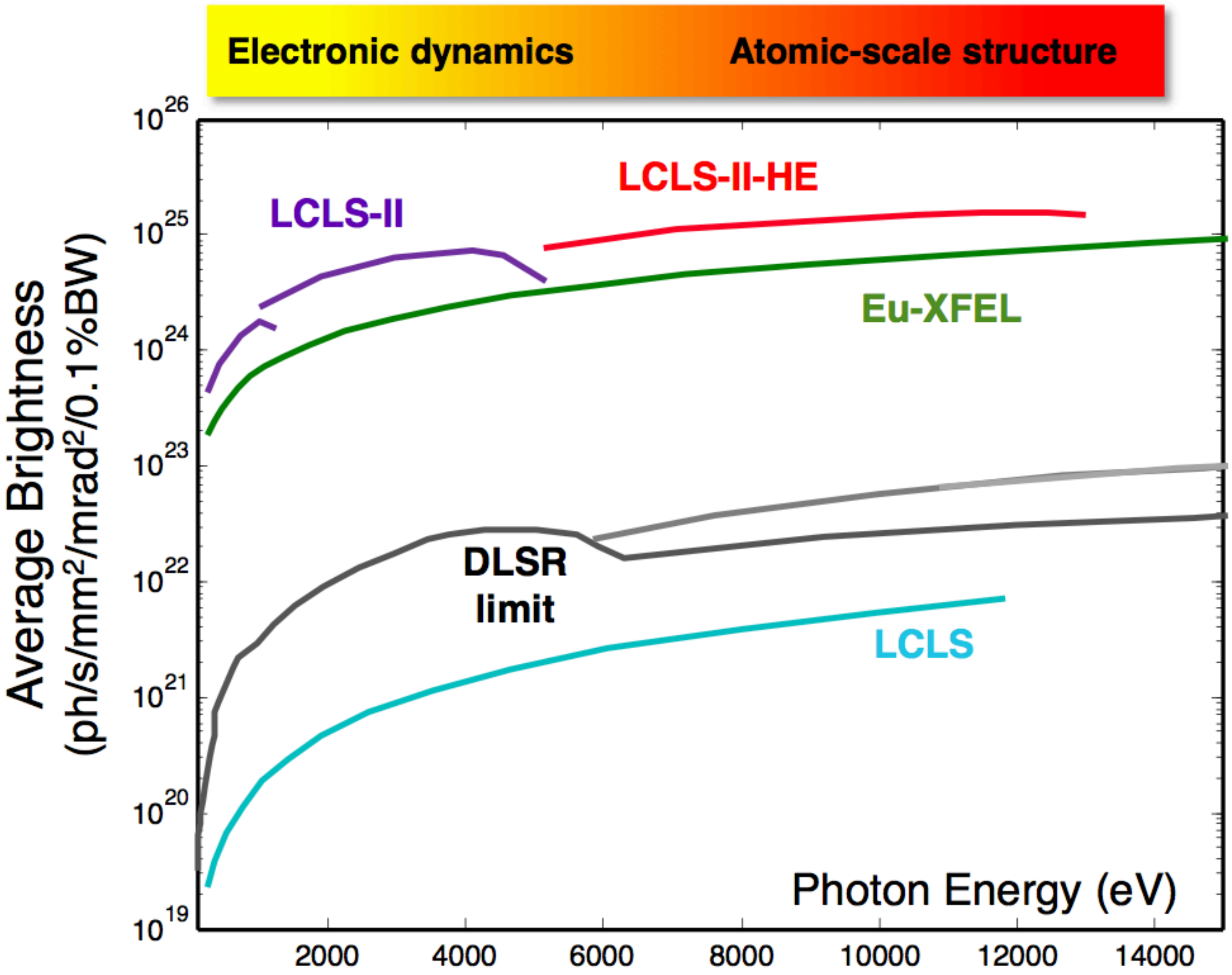
PAL-FEL, Korea
X-ray Sources: 2



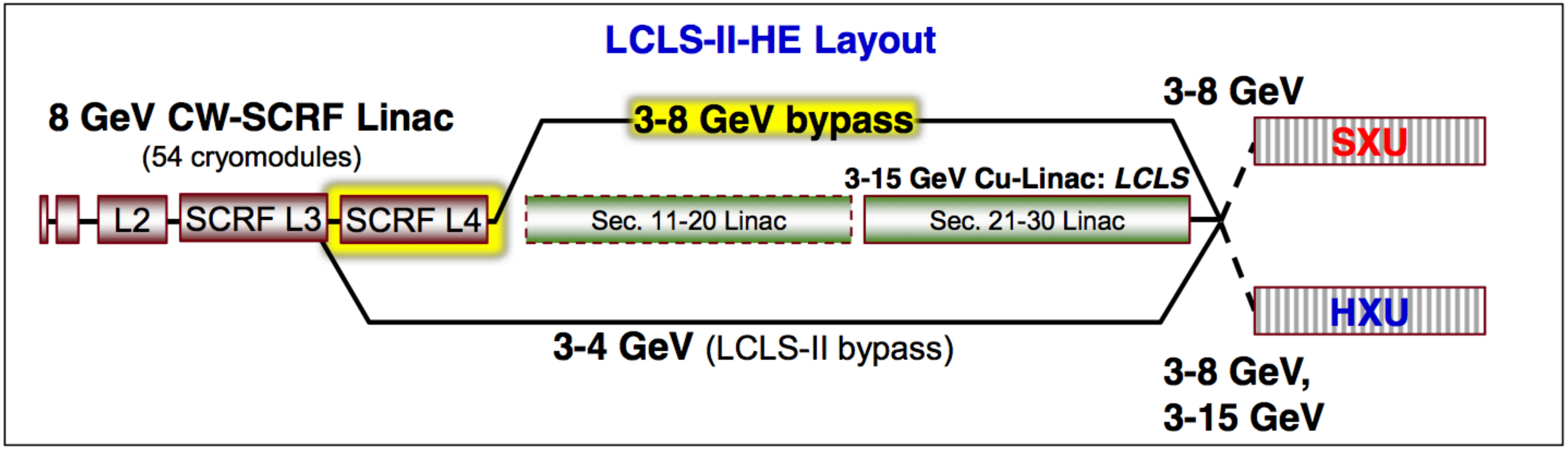
SCLF, China
X-ray Sources: 3

Shanghai XFEL, ~2025

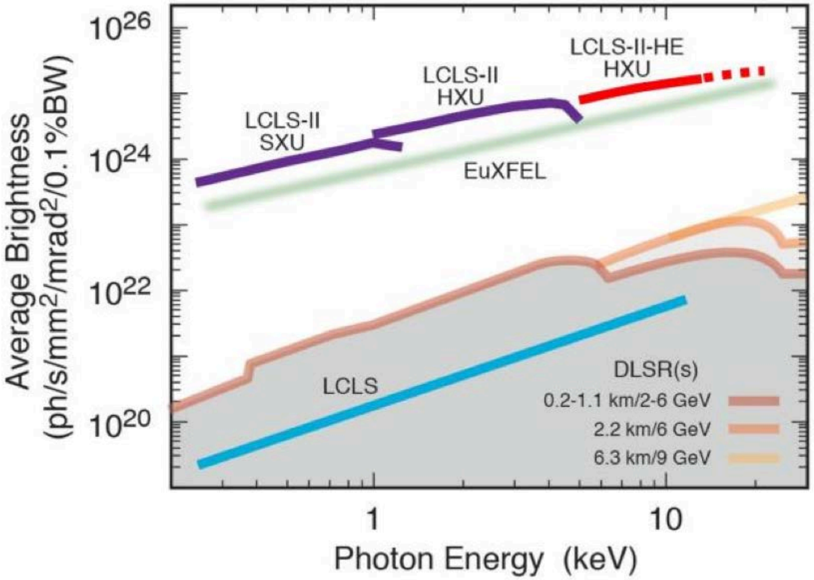
LCLS-II HE (High Energy) project was proposed (circa 2016)



Layout of LCLS-II HE allows great flexibility



Plus: existing Cu-Linac can deliver high pulse energy at 120 Hz
(to 25 keV)



Instrument Upgrade includes Dynamic X-ray Scattering

Parameter	Value
Photon energy range	10-19 keV (IXS) 4-22 keV (XPCS) (Higher energy range with harmonics)
ΔE / E	SASE, C* (111) or C* (333) mono < 10^{-6} with high resolution mono
Repetition rate	100 kHz to 1 MHz (IXS) Single pulse to >50 kHz (XPCS, detector limited)
X-ray spot size	1-500 µm
Detector arm	8 m between 0 and 55° (shorter arm for larger angle)
IXS energy resolution	3-5 meV (dependent on photon energy)
Optical laser	100 kHz, 1 mJ, OPCPA system with wavelength conversion
Temporal resolution	0.2 fs to 2 ps dependent on mode of operation
Primary X-ray techniques	Inelastic X-ray scattering X-ray photon correlation spectroscopy

Major Modifications for LCLS-II-HE

- New high resolution monochromator (in-line)
- Augment existing detector arm with inelastic scattering spectrometer
- High repetition rate detector

Science Opportunities

- Map collective excitations & understand their relation to emergent phenomena in complex materials
- Characterize materials heterogeneity, fluctuations & link to function
- High repetition rate high resolution scattering: IXS & XPCS

DXS optimized for <13 keV and capable of operating up to 19 keV

High energy-resolution photon numbers at LCLS-II-HE

- Unmatched performance from an X-ray laser (ph/s/meV)
- High spectral resolution (ph/s/meV) – far beyond DLSRs
- Time-resolution near the FT limit (200 fs ⇔ 10 meV)

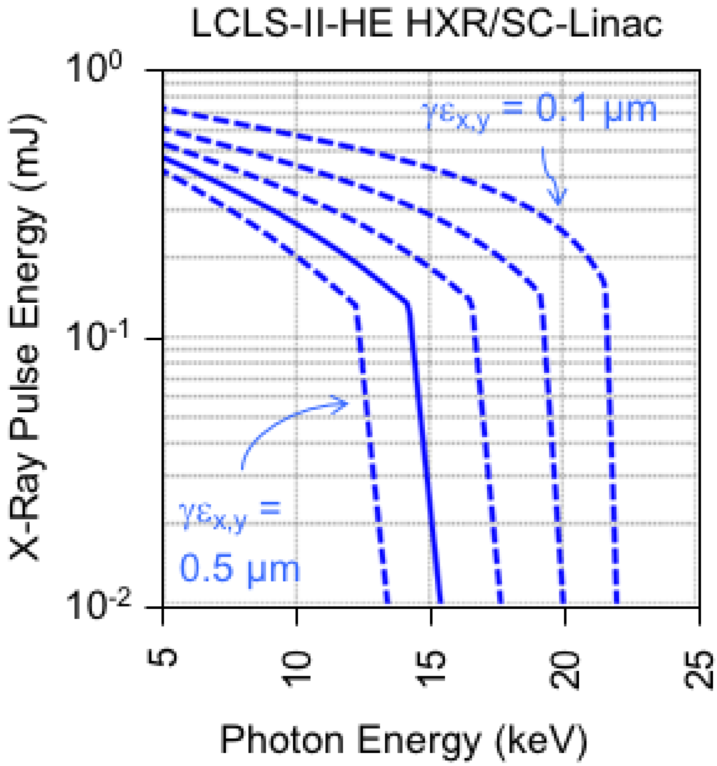
Resolution	Hard x-ray flux on sample per meV
LCLS-II-HE (seeded)	**~10^{14} ph/s**
LCLS-II-HE (SASE)	**~10^{13} ph/s**
Spring-8	~10^{11} ph/s (BL43)
ESRF	~10^{10} ph/s (ID16, ID28)
APS	~10^{10} ph/s (27-ID, 30ID) ~10^{9} ph/s (UHRIXS)
NSLS-II	~10^{10} ph/s (10-ID)

- Lower peak power (1 MHz CW) vs. EuXFEL (~30 kHz burst mode)
  - 1 µs between pulses @ LCLS-II-HE vs. 200 ns bursts @ EuXFEL
  - Approximately x10 more integrated flux (key parameter for IXS)

LCLS-II-HE received CD-1 in September 2018

SLAC NATIONAL ACCELERATOR LABORATORY	• Accelerator and FEL Design • Cryomodule and accelerator installation • Cryoplant modifications & Helium distribution installation • High Power RF, low-level RF, and Controls • X-ray instruments design & installation
Fermilab	• High Q0 & High Gradient R&D • Cryomodule design and prototype demonstration • 50% of cryomodule production • Processing for high Q • Helium distribution system design and procurement
Jefferson Lab EXPLORING THE NATURE OF MATTER	• High Q0 & High Gradient R&D • 50% of cryomodule production • Processing for high Q
CORNELL UNIVERSITY FOUNDED A.D. 1865	• High Q0 & High Gradient R&D
BERKELEY LAB	• Accelerator Physics

Develop high-brightness source is key to get to high x-ray energy

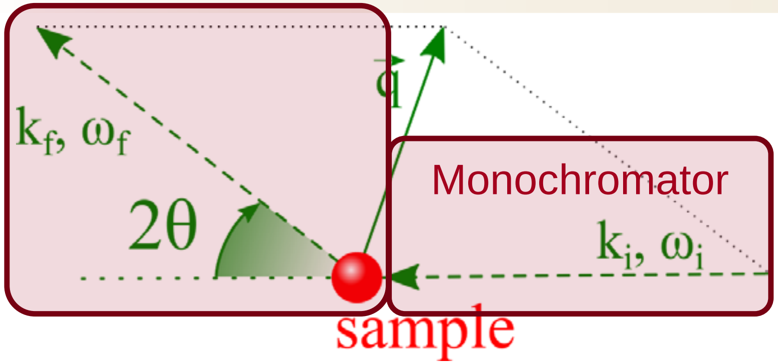


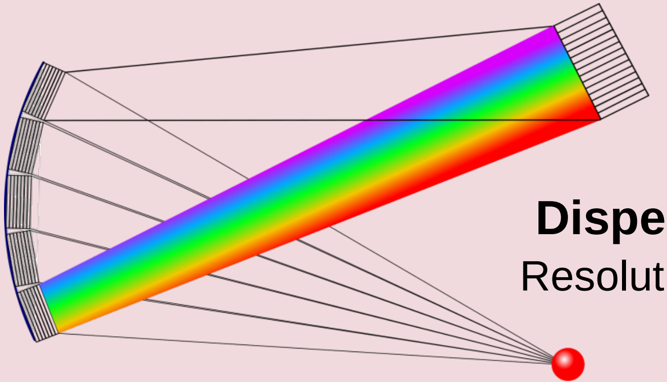
SRF Gun

High-brightness source R&D research program:

- **Cathode R&D**
  - Visible light cathode growth and characterization system for LCLS-II and beyond
- **Continuous Wave R&D**
  - High-gradient CW SRF guns for future FELs, UED/UEM and colliders
- **FEL R&D**
  - Detailed simulations of an SRF gun/injector for a new higher brightness system

Types of IXS spectrometers under consideration (Yavas)

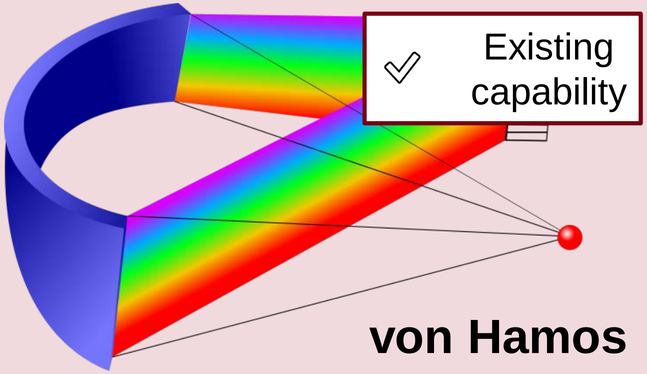




Dispersive Johann

Resolution (1-100 meV)

Option for RIXS and NIXS
Mature monochromator technology
Heat-load issue is known, analyzer needs R&D



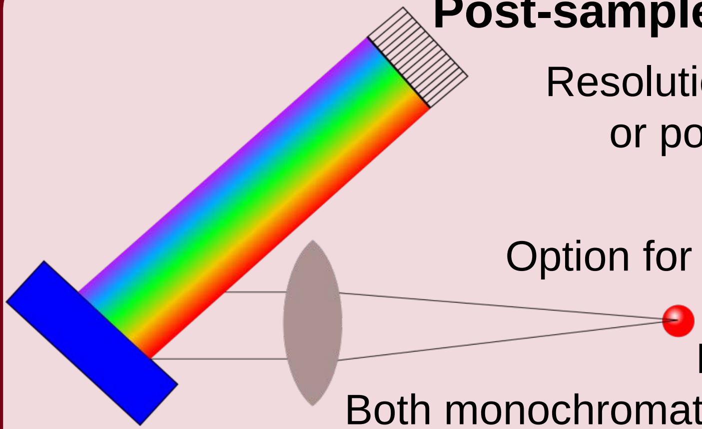
✓ Existing capability

von Hamos

for core-level excitations
Resolution (~100 meV) is limited by the analyzer geometry

Existing capability
Ready for LCLS-II-HE

Post-sample-collimation

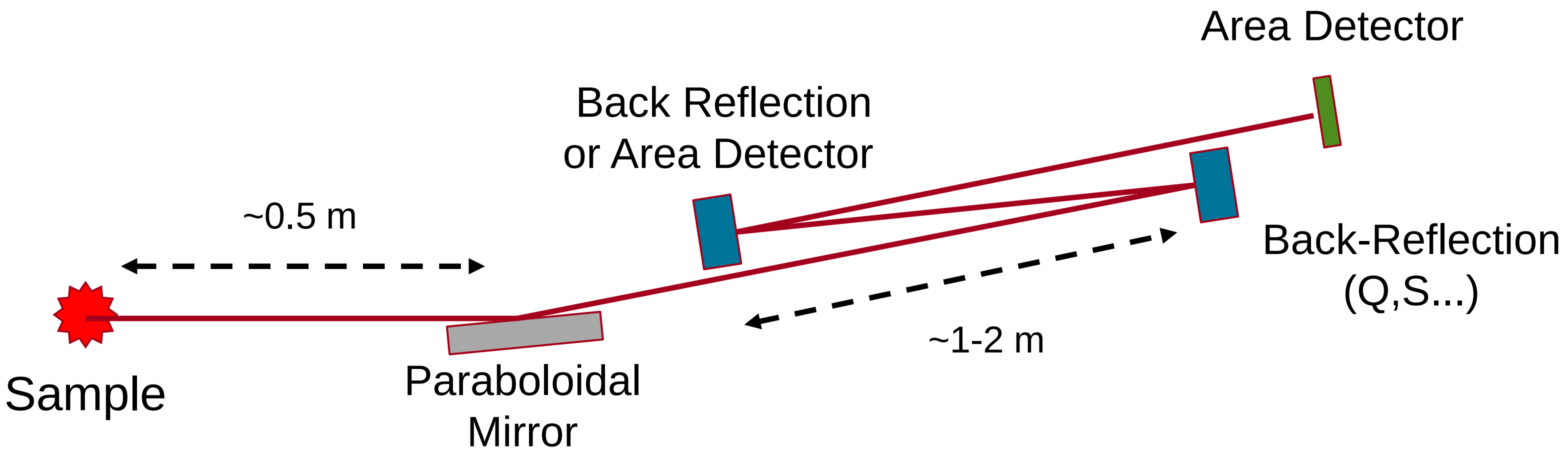


Resolution (1-10 meV) or potentially better

Option for RIXS and NIXS

New technology
Both monochromator and analyzer require R&D

One possible implementation of post-sample-collimation (Baron)



Flat Analyzers Near Backscattering:

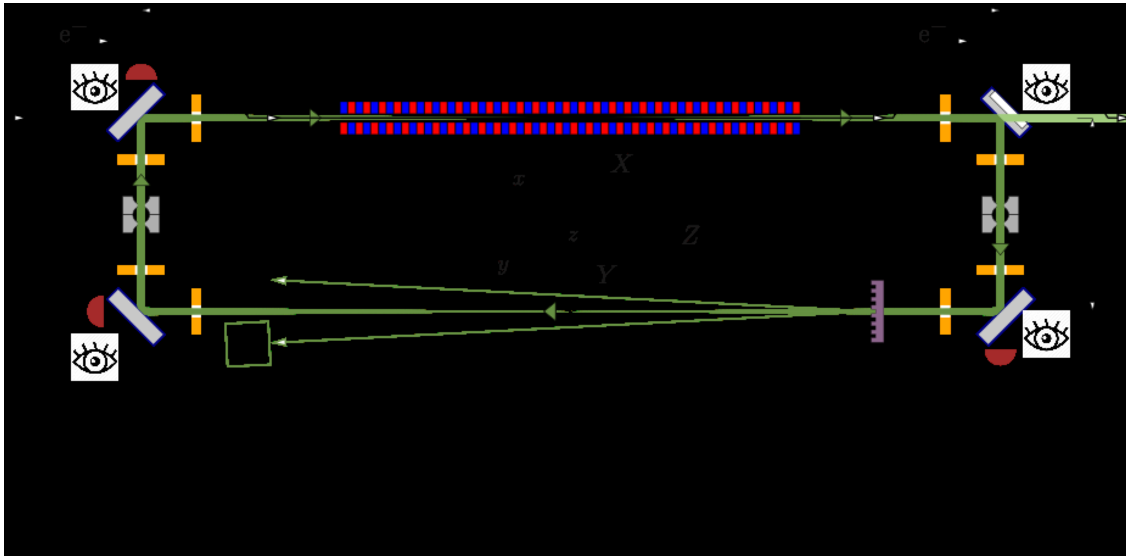
- Both RIXS (10+ meV) and NRIXS (1.2+ meV, maybe)
- Double Bounce Possible -> Good Tails ($1/E^4$)
- Easy to change crystals (and therefore resolution and operating energy)
- Mostly relaxed angular tolerances (~0.1 mrad mostly\ enough).

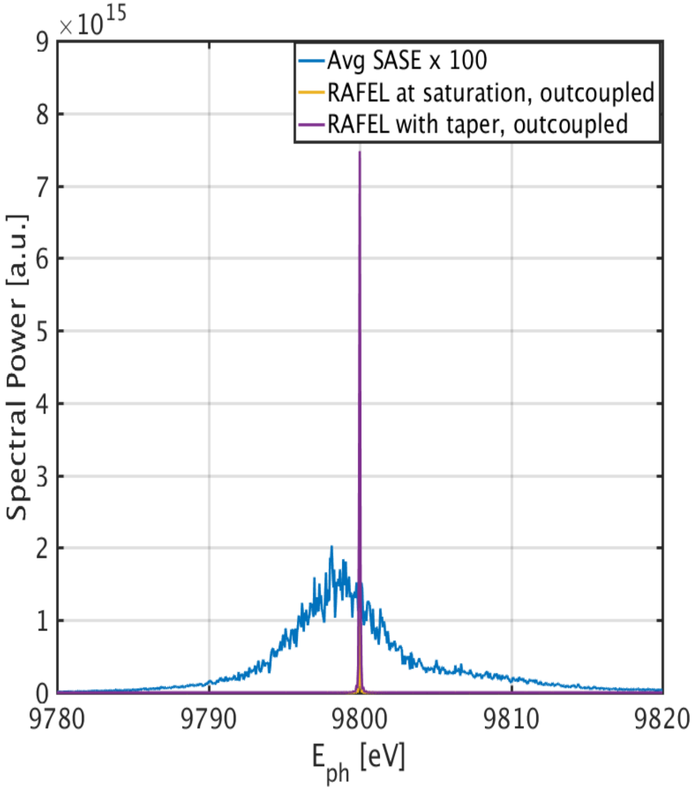
Within limits: “free” Q resolution from area detector]

[Easier collimating elements (& thicker Samples)]

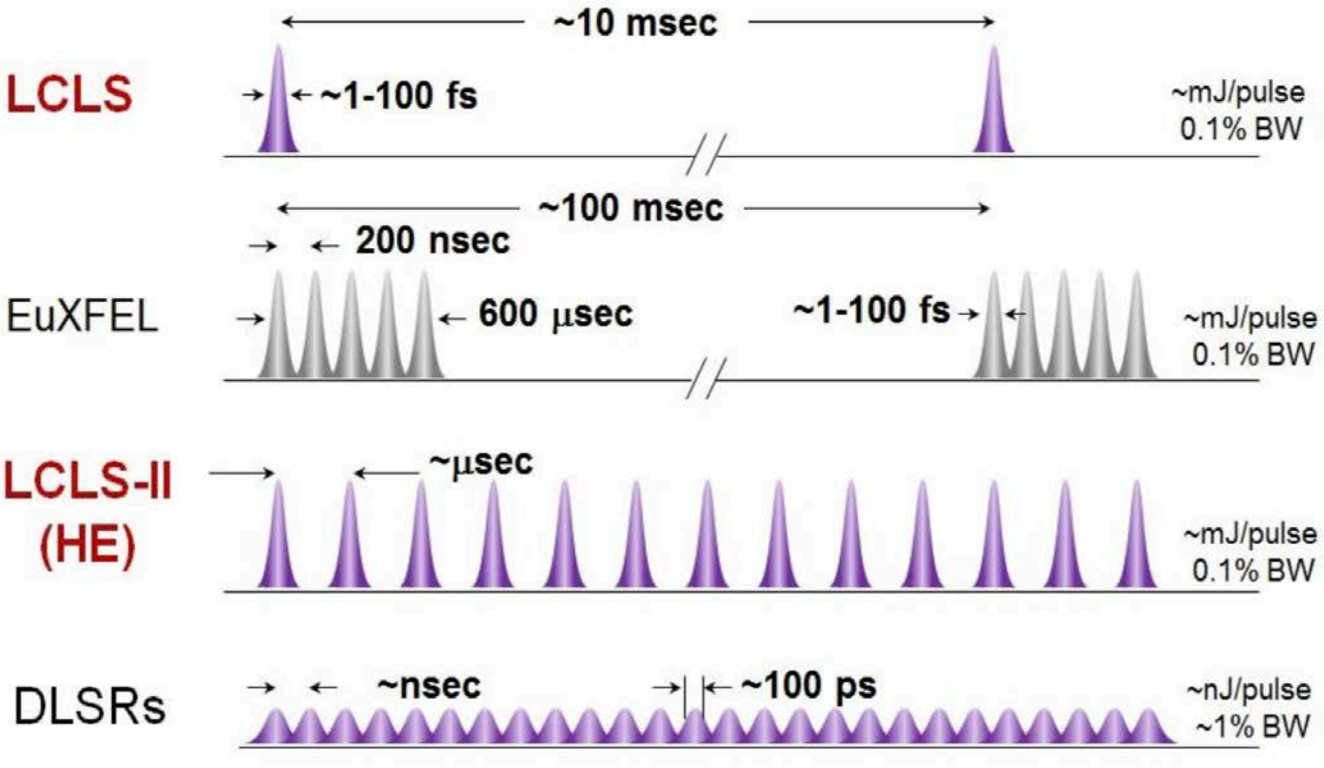
LCLS-II HE enables optical cavity-based XFELs to produce longitudinal coherent x-rays (Marcus et al; SLAC/ANL collaboration)

X-ray free electron laser oscillator (XFELO) [Kim et al] and ***X-ray regenerative amplifier free-electron laser*** (XRAFEL) [Huang et al]





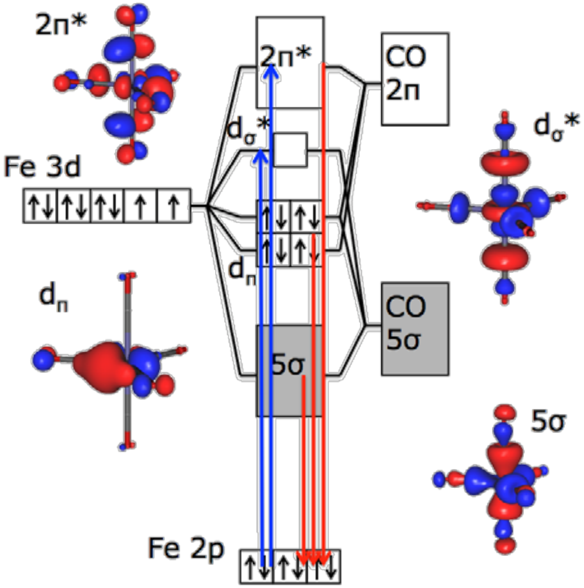
High-repetition-Rate



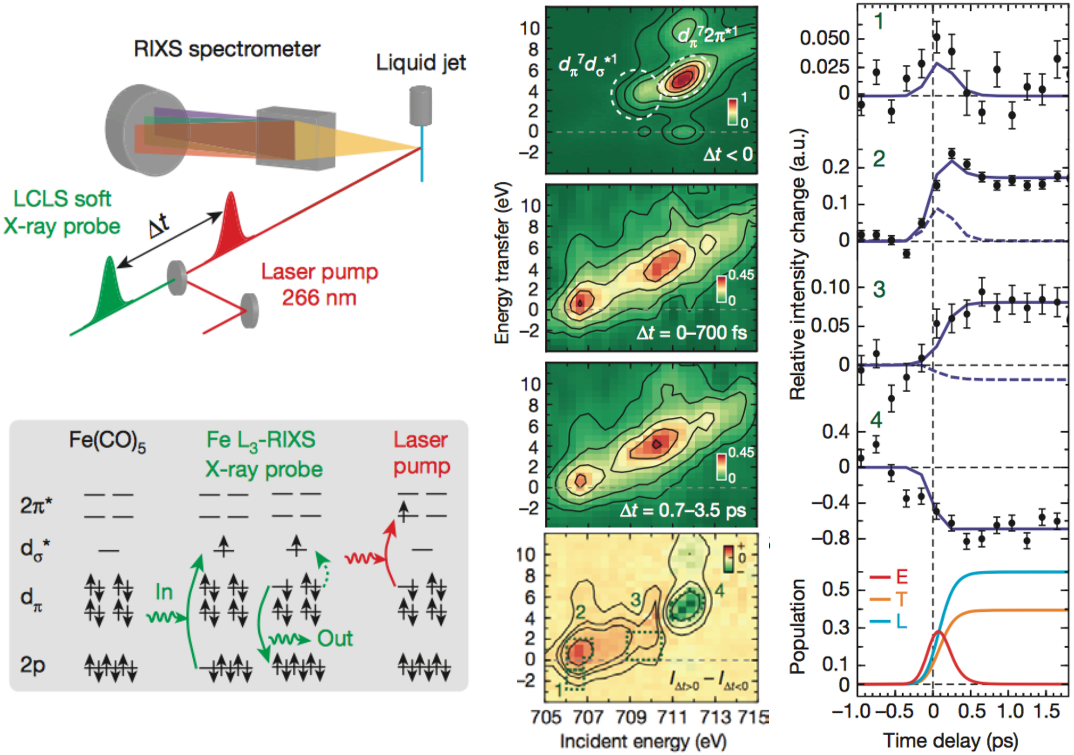
Pulse structure from LCLS warm Cu-linac at 120 Hz, burst-mode structure from the pulsed SCRF linac of the European XFEL at 5 MHz/10Hz, and the uniform (programmable) bunch structure from the CW-SCRF linac of LCLS-II-HE.

XFEL based IXS demonstration
tr-RIXS Studies of Photo-dissociation

Valence orbital structure of $Fe(CO)_5$ and the orbitals probed with RIXS at the Fe L_3 edge:



Orbital Mapping of Femtosecond Photochemistry

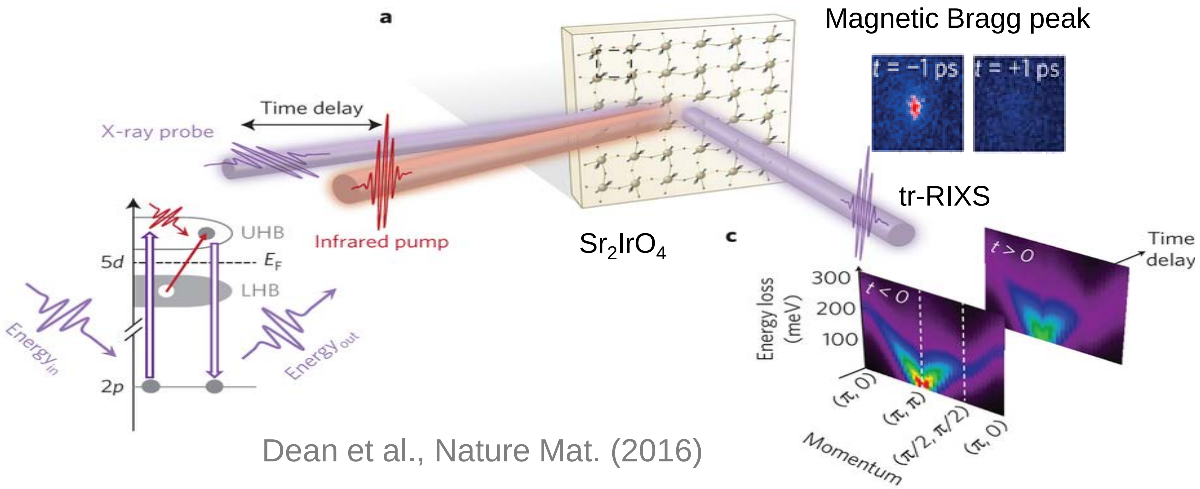


Wernet et al., Nature (2015)

With the dramatic increase in average brightness at LCLS-II, high-resolution RIXS with femtosecond resolution will be used to map how frontier orbital energies drive charge separation and transfer in complex functioning systems.

XFEL based IXS demonstration
Spin Dynamics on Sr_2IrO_4

tr-RIXS probes the magnetic quasiparticle spectrum.

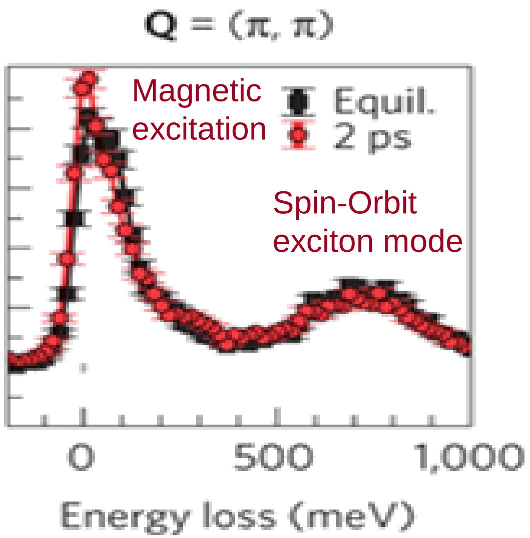


Magnetic Bragg peak

tr-RIXS

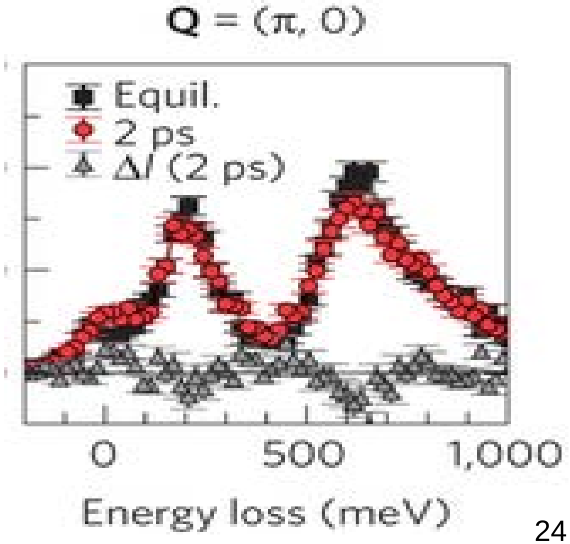
Sr_2IrO_4

Dean et al., Nature Mat. (2016)



Magnetic excitation

Spin-Orbit exciton mode

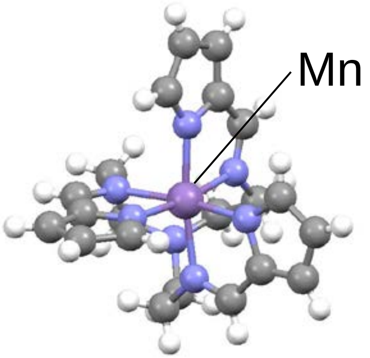


- Such ***Q***-space resolution is not available in the complementary technique of time-resolved two-magnon Raman scattering, owing to the fact that visible photons carry negligible momentum.

- This research breaks new ground by energy analyzing the scattered X-rays, that is, by performing the first ever time-resolved (tr) magnetic resonant inelastic X-ray scattering (RIXS) experiment.

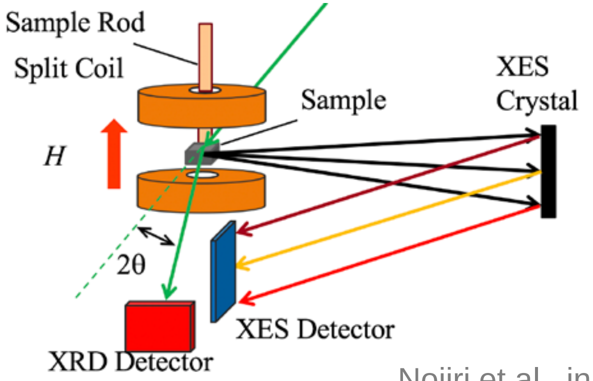
XFEL based IXS demonstration
Investigating Spin-state under Extreme-condition

Molecular structure of Mn(taa)

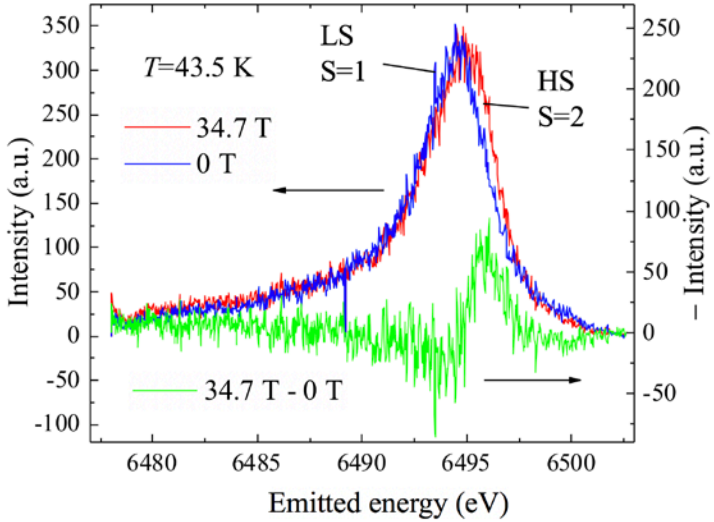


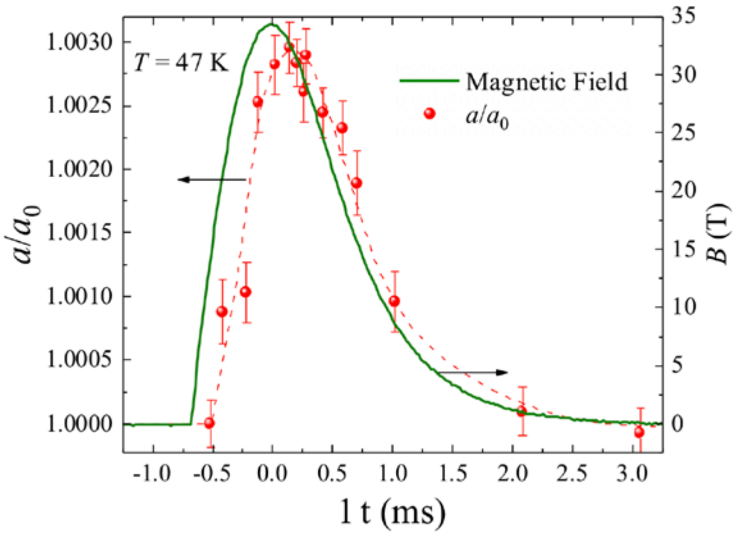
Motivation:
Exploring the expected Low-Spin (LS) to high-Spin (HS) state transition under a high magnetic field

Simultaneous XRD&XES measurement



Nojiri et al., in preparation



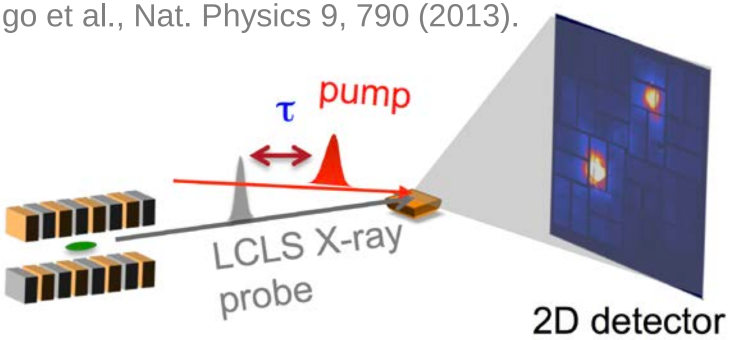


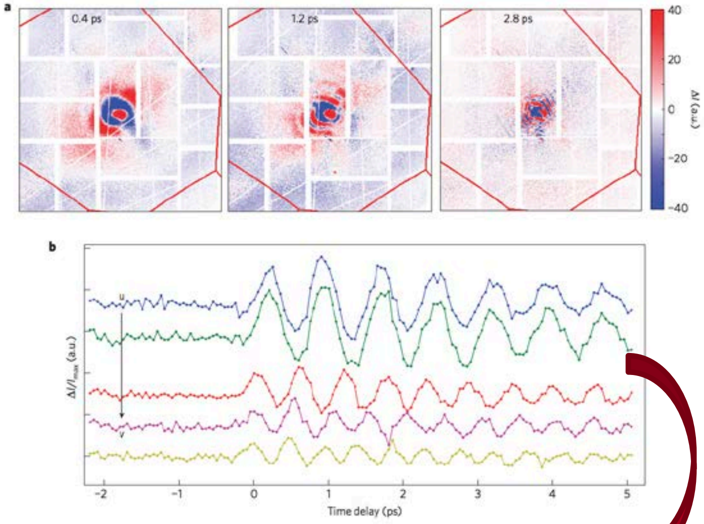
By using synchronizing a high magnetic pulse and x-ray pulse, the spin transition (LS ➔ HS), its mechanism "dynamic Jahn-Teller distortion" were directly observed.

XFEL based IXS demonstration
Phonon spectroscopy by Fourier-Transform IXS

Fourier Transform (FT) Inelastic X-ray Scattering at FELs

Trigo et al., Nat. Physics 9, 790 (2013).

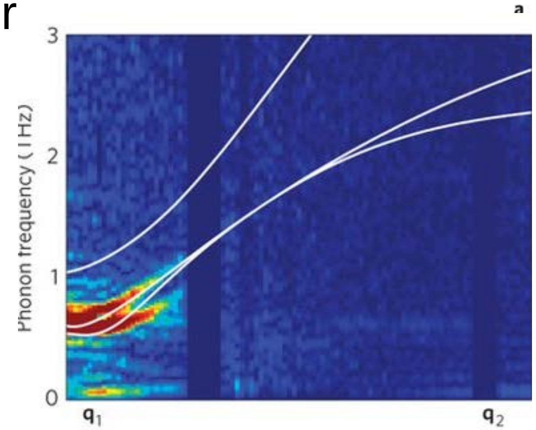


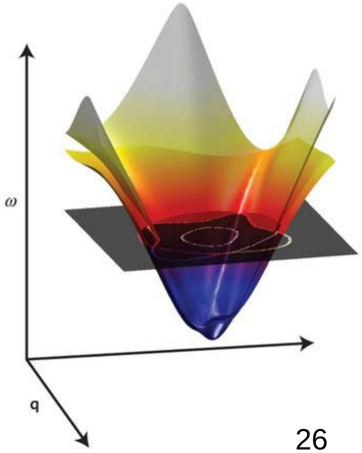


FT

The time-domain measurements are a direct way to follow the excitations of solids and the flow of energy well away from their "home" positions and ground state.

This x-ray scattering technique allows us to track the motion of atoms as they respond to sudden changes in their energy state.





Summary

- IXS has come a long way
- It benefits and pushes the state-of-art accelerator, instrumentation, and theory
- The technique has brought spectroscopy, scattering and now ultra-fast and accelerator community together
- The next decade is going to be even more exciting as DLSR and high rep. rate FELs come online

Acknowledgement

SLAC:

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Hasan Yavas

Bob Schoenlein

Georgi Dakovski

Greg Hays

Gabriel Marcus

Zirong Huang

SPring-8/RIKEN:

Alfred Baron

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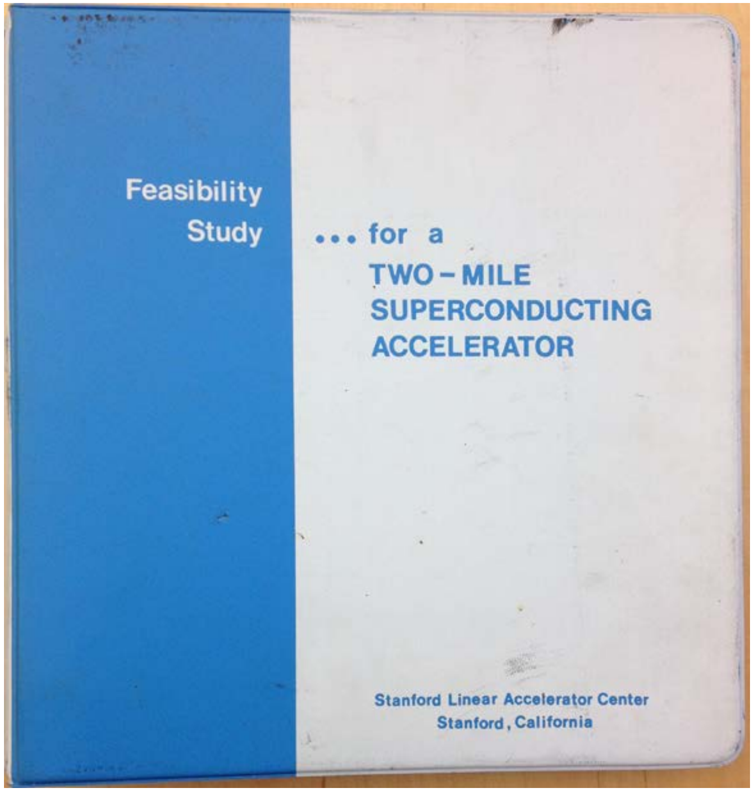


Feasibility Study

... for a TWO-MILE SUPERCONDUCTING ACCELERATOR

Stanford Linear Accelerator Center
Stanford, California

X-ray Raman Scattering & (Resonant) IXS

Method	Applications	Energy (eV)	Resolution
XRS*	Core-level excitations	6000 – 13000	~500 meV
RIXS**	Charge, orbital & spin excitations	7000 – 12000	<10 – 100 meV
NIXS***	Lattice excitations	9000 – 25000	1 – 6 meV

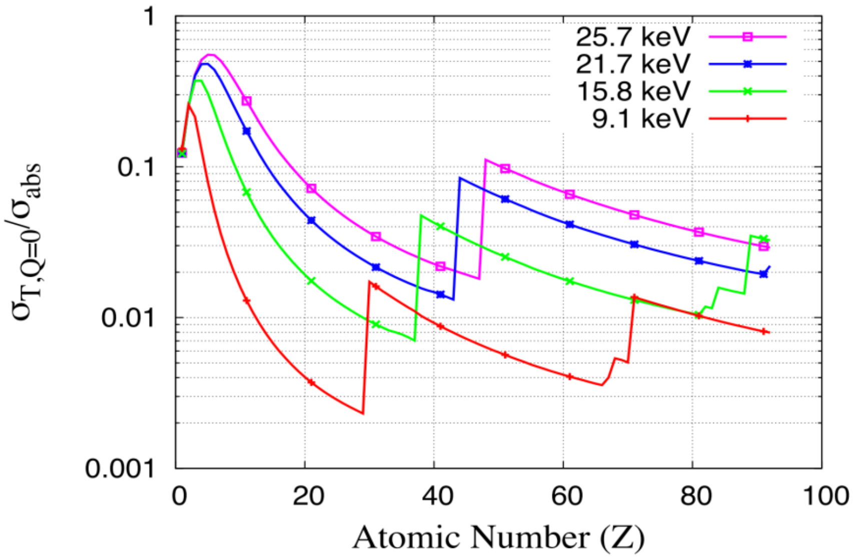
✓ Existing capability

New Development

* SSRL, APS, ESRF, Spring8, PETRA-III, Soleil
** APS, ESRF, Spring8
*** APS, ESRF, NSLS-II, Spring8

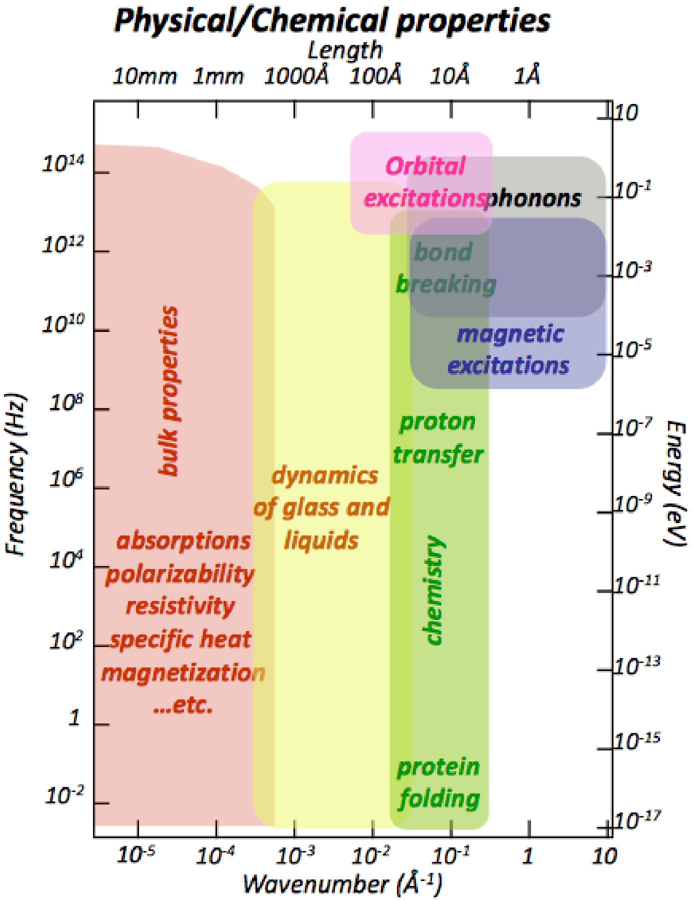
Motivation for higher energy photons
For non-resonant scattering,
a typical high-Z material would yield
x4 less signal at 12 keV compared to 20 keV
(identical number of photons & resolution)

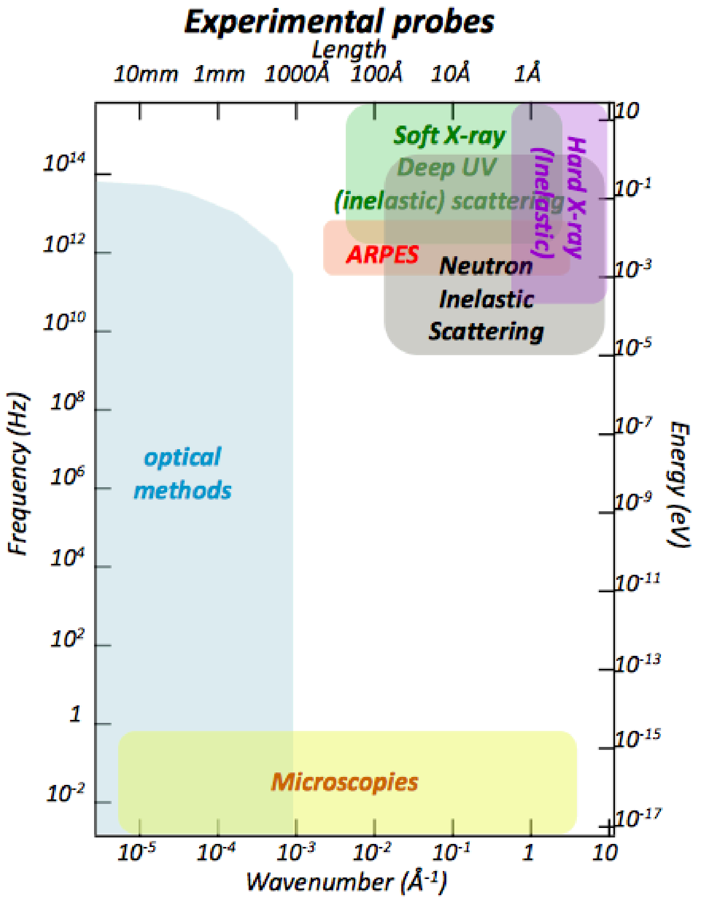
LCLS-II-HE parameters can make it up



Selection of experimental tools

Figures' courtesy of Yi-De Chuang





Inelastic x-ray scattering is the ideal tool for exploring many exotic properties in materials.

Inelastic x-ray scattering (IXS): *Scientific Information*

- **Atomic Dynamics**

➔ Monitoring phenomena of phonons in a solid (or liquid)

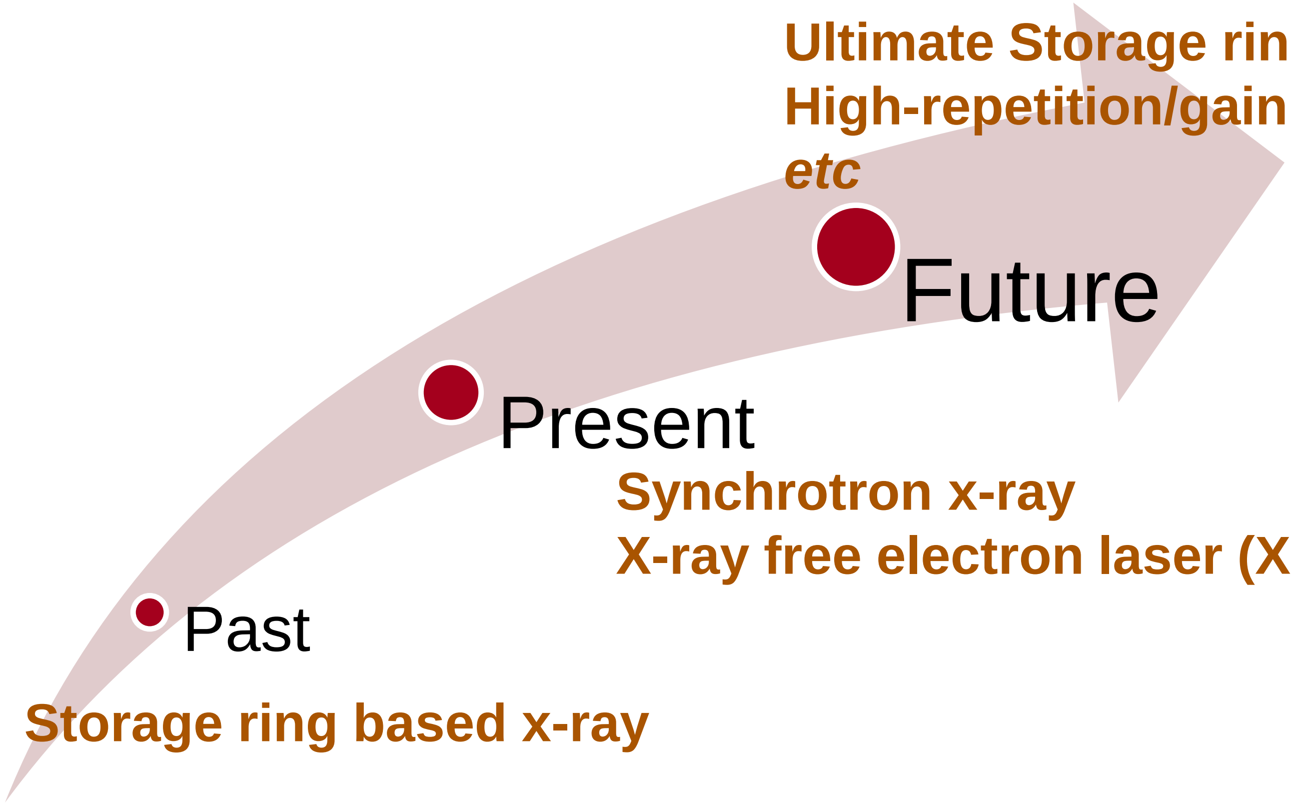
Advantage in *Non-resonant*	A high resolution: Choose energy to match optics

- **Electronic Dynamics**

➔Monitoring phenomena of electron transition (excitations)

Advantage in *Resonant*	Tunable an atomic transition energy (absorption edges)

X-ray sources



Ultimate Storage ring (USR)
High-repetition/gain FEL (e.g. LCLS-II)
etc

Future

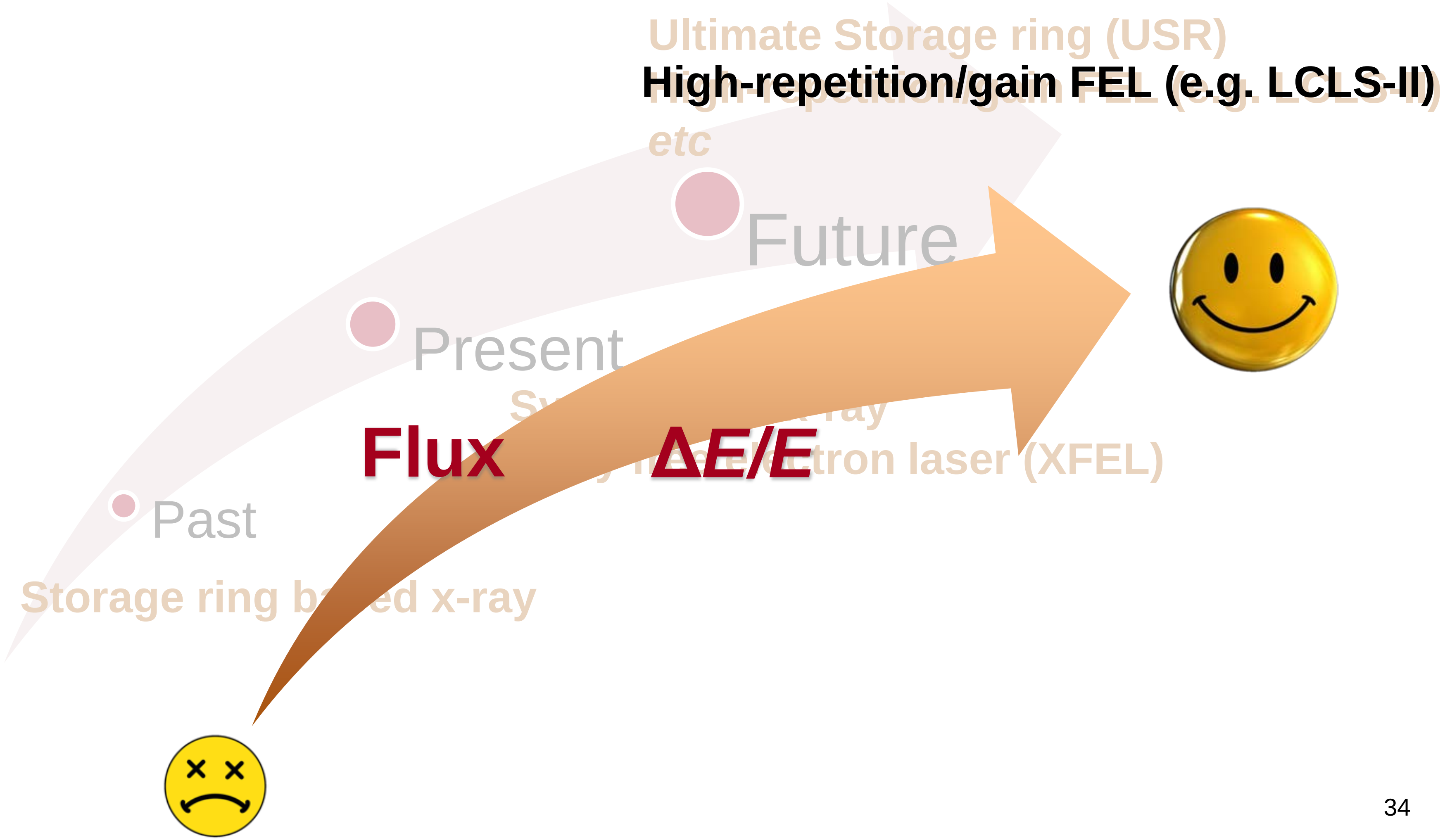
Present

Synchrotron x-ray
X-ray free electron laser (XFEL)

Past

Storage ring based x-ray

X-ray sources for IXS



Past

Storage ring b… ed x-ray

Present

Sy… -ray

…y free ele…ctron laser (XFEL)

Future

Ultimate Storage ring (USR)

High-repetition/gain FEL (e.g. LCLS-II)

etc

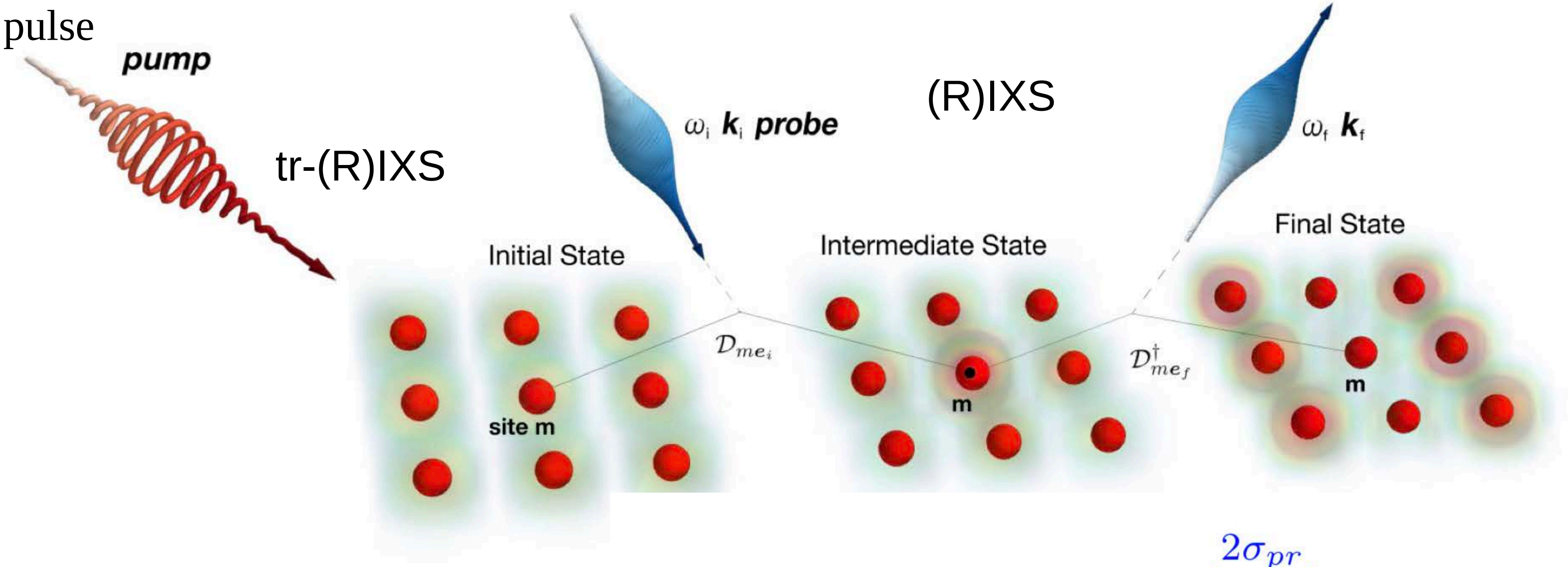
Flux

Δ*E*/*E*

XFEL based IXS demonstration

Chen et al., PRB **99**, 104306 (2019)

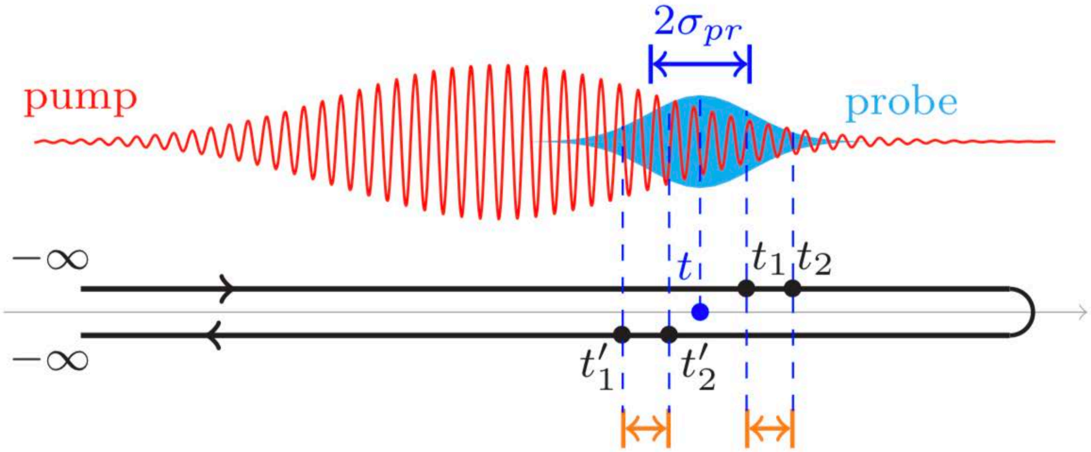
perturbation via the pump pulse



tr-(R)IXS

(R)IXS

Non-equilibrium physics in solids and its relaxation to equilibrium state is a largely unexplored region.



$\Delta E/E$

Flux

Coherence

Time

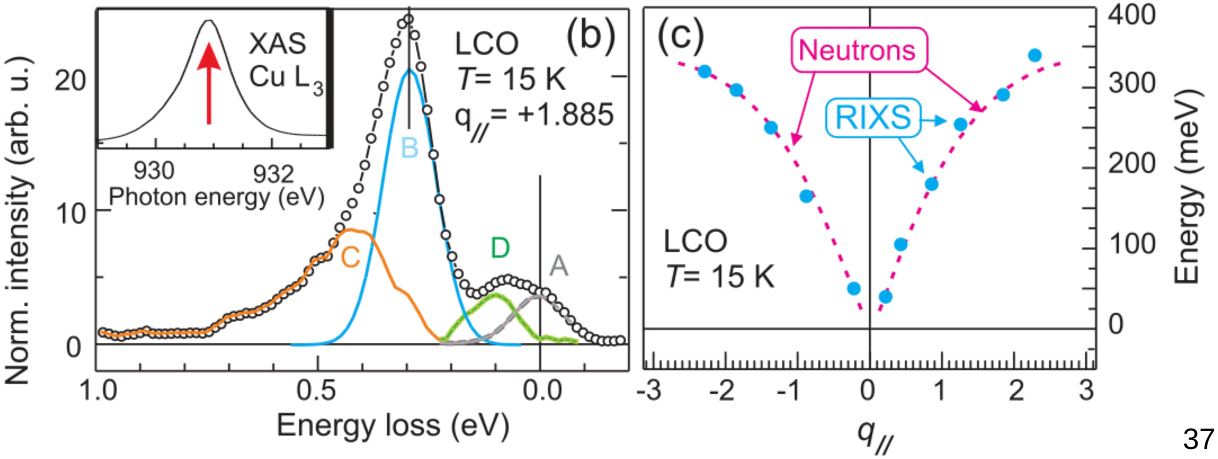
Benefit of soft x-ray

Although the finite momentum transfer could be an issue, due to their advantage, it can be helpful in exploring the dispersion of excitations;

edge	Energy range	Resolving power for 10 meV	Advantage	Disadvantage
L	500 ~ 1000 eV	1 keV/10 meV = 10^5	-Large ΔQ -Small elastic peak on resonance -Fewer branching (multiplets)	-Large machine -Lower throughput -expansive
M	50 ~ 100 eV	100 eV/10 meV = 10^4	-Small ΔQ -Smaller machine -High throughput	-Strong elastic peak -Complex branching

Huge advantage in High-Tc Cuprates

e.g., Single magnon dispersion probed by IXS and neutron



Braicovich et al., PRL (2010)

Plan for soft x-ray instrumentation

High-resolution tr-RIXS setup

High-resolution RIXS setup at soft x-ray

Need an input

Plan for hard x-ray instrumentation
Medium energy resolution (short term)

- **An IXS instrument that can be utilized for**

  Ir L_3 edge (Resonant): (4 meV is available today)
    - 90 degrees scattering angle for polarization

  Non-resonant IXS for phonons:
    - 1-3 meV overall resolution
    - Up to ~135 degrees two-theta
    - No polarization flip

- **11.2 keV, FT-limited matching energy-resolution pulses**

- **Small pixel size detectors (<25 micron approaching 1 micron)**

- **Integrated sample environment (Low Temperature, Magnetic field, diamond anvil cell, etc.)**

Plan for hard x-ray instrumentation

Medium energy resolution (short term)

Instrumentation home: XCS hutch (tentative)

Beyond LCLS-II HE

Optical cavity-based X-ray free electron laser

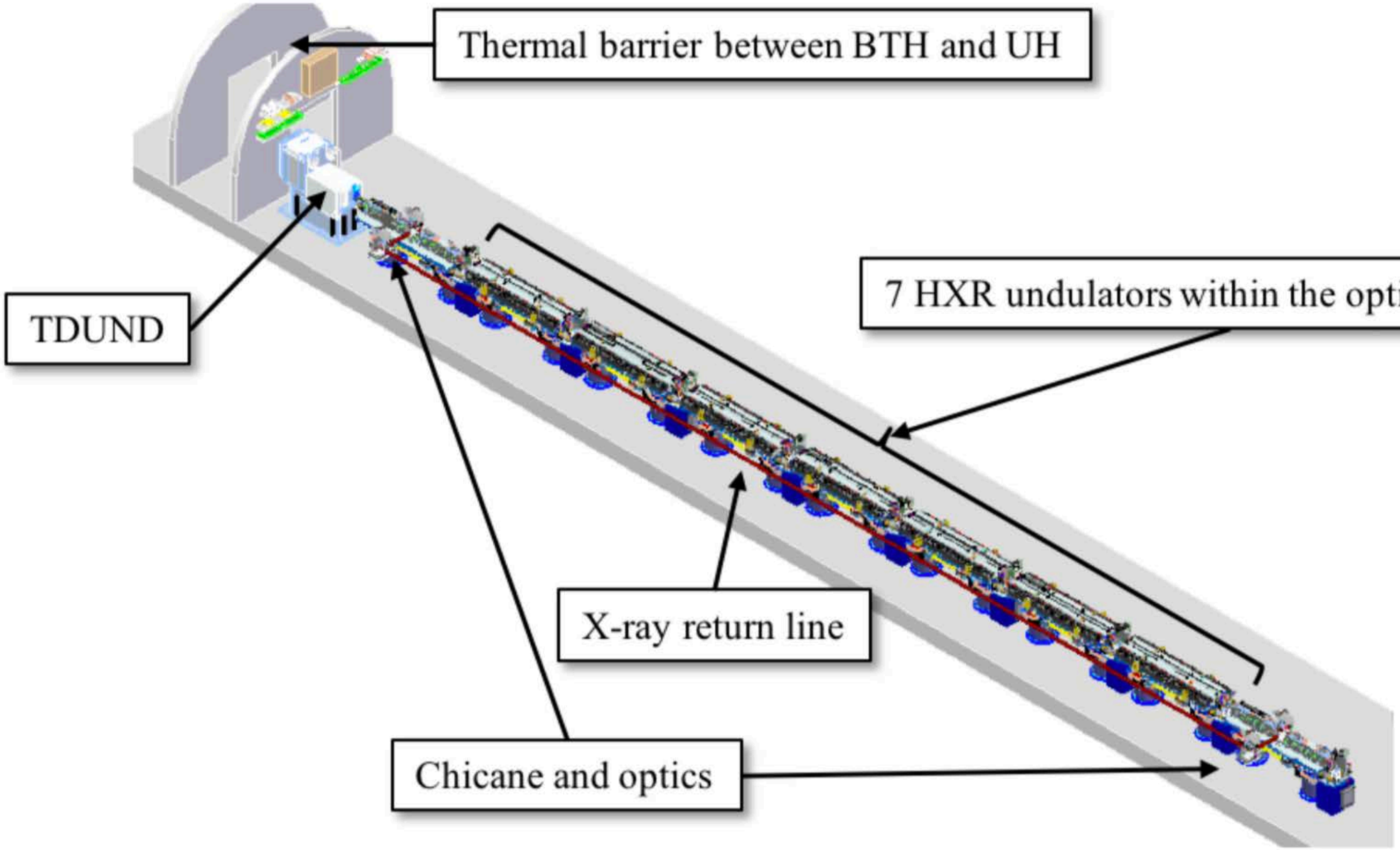
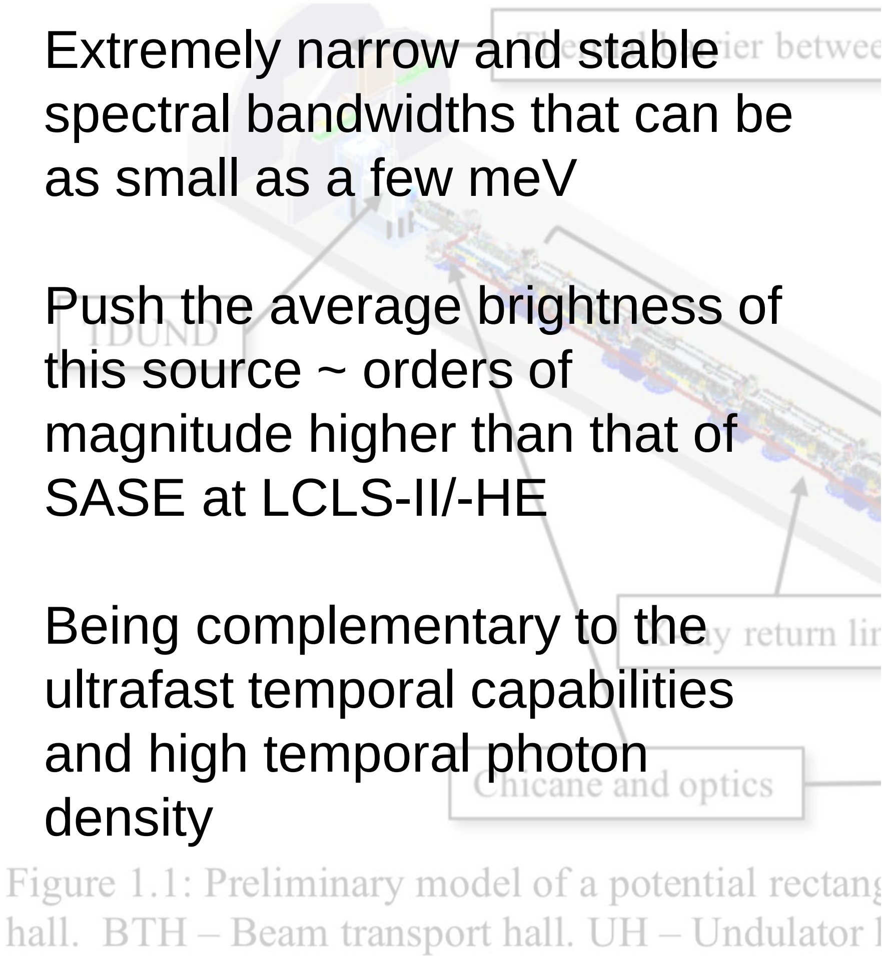


Figure 1.1: Preliminary model of a potential rectangular X-ray cavity in the LCLS-II undulator hall. BTH – Beam transport hall. UH – Undulator hall. TDUND – Undulator tune-up dump.

Beyond LCLS-II HE: Regenerative

: Optical cavity-based X-ray free electron laser



- Extremely narrow and stable spectral bandwidths that can be as small as a few meV

- Push the average brightness of this source ~ orders of magnitude higher than that of SASE at LCLS-II/-HE

- Being complementary to the ultrafast temporal capabilities and high temporal photon density

Figure 1.1: Preliminary model of a potential rectang
hall. BTH – Beam transport hall. UH – Undulator l

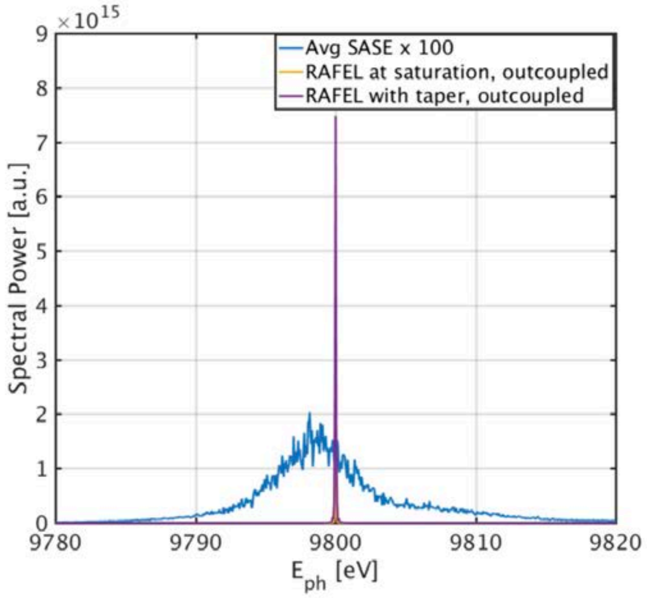


Figure 3.2: LCLS-II-HE spectral comparison at a 9.8 keV photon energy using a 100 pC electron beam for tapered SASE x 100 (red – individual shots, blue - average), XRAFEL at saturation (yellow) and tapered XRAFEL (purple). The peak spectral brightness of saturated (tapered) XRAFEL is 75 (550) times larger than average SASE.

Beyond LCLS-II HE

Optical cavity-based X-ray free electron laser

Since this proposed new source is possible to deliver X-rays with high peak power at high repetition rate, as well as high coherence, it could directly affect IXS's instrumental/experimetnal limits.

Figure 6.7: Top – The LCLS-II HXR undulator configuration. From left to right: TDUND, 7 HXR undulators, empty section for 2-stage hard X-ray self-seeding, following undulators. Bottom – Preliminary mockup of the proposed HXR undulator configuration including the rectangular X-ray optical cavity (sans in-situ diagnostic line). From left to right: TDUND, chicane and optics chamber, 7 HXR undulators, chicane and optics chamber. The red line illustrates the X-ray cavity return line.

Summary

NOTE: Please put your summary/message after finalizing your slide.